Power dependence of upconversion luminescence

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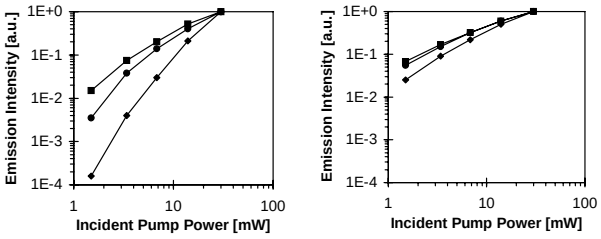
Spectroscopic data are of essential value for understanding the excitation mechanisms in luminescent and laser materials. Special attention has been devoted to the investigation of upconversion-induced luminescence, partly because of the availability of near-infrared pump sources for the excitation of visible luminescence and laser emission [1] and partly because these mechanisms can introduce a loss channel for devices emitting in the infrared region [2,3]. Typical upconversion processes are pump excited-state absorption (ESA) and energy-transfer upconversion (ETU). For the interpretation of short-wavelength luminescence, it is often assumed that the order *n* of the upconversion process, i.e. the number *n* of pump photons required to excite the emitting state, is indicated by the slope of the luminescence intensity versus pump power in double-logarithmic representation. However, a saturation of upconversion luminescence with rising pump power was already observed 30 years ago [4].

We show theoretically by assuming the simplest possible rate-equation model that the experimentally observed decrease in power dependence of an upconversion luminescence with rising pump power is purely determined by the competition between linear decay and upconversion processes for the depletion of the intermediate excited states. An upconversion luminescence which is excited by the subsequent absorption of *n* photons has a dependence on absorbed pump power *P* which may range from P^n in the limes of infinitely short lifetimes of the intermediate excited states down to P^1 for the upper state and less than P^1 for the intermediate states in the limes of infinitely strong upconversion rates. Whereas the upper limes is valid universally, the lower limes for the intermediate states depends on whether the excitation is achieved via ETU or ESA and whether the states decay predominantly via ground-state luminescence or multiphonon relaxation into the next lower-lying state. These results allow for the interpretation of a measured power dependence of multiphoton-excited luminescence with respect to the order of the process and its physical origin and strength.

We have verified experimentally the convergence into both limes by upconversion luminescence in $Cs_3Lu_2Cl_9$:1% Er^{3+} and $Cs_3Er_2Cl_9$ [5], see Figures.



Figs.: Power dependence of upconversion luminescence [5] from $^4I_{9/2}$ (■), $^4S_{3/2}$ (●), and $^2H_{9/2}$ (◆) in $Cs_3Lu_2Cl_9$:1% Er^{3+} (left) and $Cs_3Er_2Cl_9$ (right). The Er^{3+} ions were excited at 1.54 µm into the $^4I_{13/2}$ level. The power dependence ranges from P^n (left, ETU small) to $P^{1/2}$ for $^4I_{9/2}$ and $^4S_{3/2}$ and P^1 for $^2H_{9/2}$ (right, ETU large).

References

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