Spontaneous transitions to enhanced fluorescence for GeV centers in a single microcrystalline diamond

Natalia A. Lozing[1,2,*], *Maxim G.* Gladush[1,3], *Ivan Yu.* Eremchev[1], *Eugeniy A.* Ekimov[4], and *Andrey V.* Naumov

[1]Institute for Spectroscopy RAS, 5 Fizicheskaya Str., Troitsk, Moscow 108840, Russia
[2]National Research University Higher School of Economics, 20 Myasnitskaya Str., Moscow 101000 Russia
[3]Moscow State Pedagogical University,1/1 M. Pirogovskaya Str., Moscow 119991, Russia
[4]Vereshchagin Institute for High Pressure Physics of the Russian Academy of Sciences, 14 Kalushskoye Shosse, Troitsk, Moscow 108840, Russia

Abstract. We propose a theoretical explanation of spontaneous transitions between dim and bright fluorescence intensity states observed experimentally in a microcrystal of diamond with germanium-vacancy colour centres driven by a continuous wave laser. We use a generalized system of optical Maxwell-Bloch equations derived for an emitter in an ensemble of motionless similar particles embedded in a dielectric medium, which is transparent for the incident light. A numerical analysis of transient regimes and several models of slow damping of the bright luminescence mode are reported.

Spontaneous flickering of the fluorescence intensity was registered experimentally for a microcrystal of diamond with germanium-vacancy centres excited by a continuous wave laser light. The fluorescence intensity from the sample was observed to be switching to several bright modes and returning to the only well determined dim mode at random occasions on the time-scale of seconds. The effect demonstrated smooth transition dynamics on the same scale and tended to last for hours. The phenomenon had a strict power threshold. These characteristics are far from the effect of "blinking" but are similar to experiments in micron-sized crystals of methylammonium lead bromide and CaS:Eu2+ crystals [1-3]. It can be assumed that the observed effect is likely to be due to nonlinear interactions of the incident light with the photoactive sites inside the crystal. It is known that the optical properties of dense atomic ensembles or complex materials can differ greatly from the optical properties exhibited by independent quantum emitters. In this report we propose a theoretical explanation based on the possibility for a cooperative ensemble of emitting centres to show intrinsic cooperative optical multistability [4]. It is shown that the Maxwell-Bloch type equations describing such a system [5], even in the presence of large values of the phase relaxation rate, retain the possibility of having several stable steady state solutions that form a multistable optical response for certain

* Corresponding author: lozing@isan.troitsk.ru

combinations of the pump power and concentration of the emitters. A numerical analysis has shown that the transition time may exceed the lifetime by an order of magnitude. We report several models of slow damping of the bright fluorescence mode and discuss alternative reasons for such a behaviour.

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