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INFLUENCE OF DEPTH-DEPENDENT EXCITATION-LIGHT ABSORPTION ON PHOTOLUMINESCENCE EMISSION FROM A SLAB

RT/2025/16/ENEA

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E. Nichelatti

Abstract
This Technical Report investigates how the absorption of excitation light (optical pump) affects photoluminescence (PL) intensity measurements in solids with parallel planar faces containing a depth-dependent distribution of emitting centers. A correction method based on optical density measurements is proposed to account for the gradual attenuation of the pump light across the material thickness. In the specific case of color centers generated by ionizing radiation in LiF crystals, the study highlights the possible systematic errors introduced by pump absorption in estimating the saturation dose, assuming it is derived from best fits of PL intensity as a function of absorbed dose. Several examples are presented to illustrate how PL trends are influenced not only by pump absorption, but also by the spatial distribution of the absorbed dose within the crystal. The findings emphasize the importance of correcting for pump absorption when evaluating PL saturation behavior in irradiated LiF crystal slabs.

Keywords: *photoluminescence correction, optical pump absorption, color centers in LiF, saturation dose estimation, ionizing radiation effects.*

Sommario
In questo Rapporto Tecnico viene analizzato come l'assorbimento della luce di eccitazione (pompa ottica) influenzi le misure di intensità di fotoluminescenza (FL) in solidi a facce piane parallele contenenti una distribuzione di centri emettitori variabile lungo lo spessore. Viene proposta una metodologia di correzione basata sulla misura della densità ottica, per tenere conto dell'attenuazione progressiva della luce di pompa attraverso il materiale. Nel caso specifico di centri di colore generati da irraggiamento ionizzante in cristalli di LiF, lo studio evidenzia i possibili errori sistematici introdotti dall'assorbimento della pompa nella stima della dose di saturazione, assumendo che quest'ultima venga ottenuta tramite best fit degli andamenti di intensità di FL in funzione della dose assorbita. Vengono infine presentati alcuni esempi che mostrano come tali andamenti siano influenzati non solo dall'assorbimento della pompa ottica, ma anche dalla distribuzione spaziale della dose assorbita all'interno del cristallo. I risultati sottolineano l'importanza di correggere l'effetto di assorbimento della pompa nella valutazione del comportamento di saturazione della FL in cristalli di LiF irradiati.

Parole chiave: correzione della fotoluminescenza, assorbimento della pompa ottica, centri di colore nel LiF, stima della dose di saturazione, effetti della radiazione ionizzante.

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1. Introduction

Typically, when measuring the photoluminescence (PL) intensity from optically active centers embedded in a material, an electromagnetic excitation wave (optical pump) impinges on the material and drives the so-called optical cycle of the centers, which consists of the absorption of pump photons and the emission of Stokes-shifted, lower-energy (larger-wavelength) photons. The emitted photons are then collected by an optical detector. If, in order to excite the emitting centers, the optical pump must traverse a layer of material that also contains such centers and therefore gradually loses photons as it propagates, it is evident that its intensity will progressively decrease relative to its initial value upon entering the material. In other words, when the volume containing the centers to be optically excited is not negligibly thin, the pump intensity within it is not uniform, but decreases with the thickness of the material traversed.

This Technical Report aims to calculate, within the limits of a few simplifying assumptions, the PL intensity produced by a volume containing emitting centers whose density along the thickness—coinciding with the propagation axis of the optical pump—follows an arbitrary spatial distribution. Additionally, a method is presented to correct the distortions introduced by pump absorption in PL intensity measurements using optical density data. Finally, for emitting centers generated by ionizing radiation, specific examples of spatial dose distributions are provided to illustrate how the estimation of the material's saturation dose is influenced by pump absorption effects.

2. Theory

2.1 Propagation and absorption of the optical pump

The optical pump is assumed to be a plane, monochromatic electromagnetic wave propagating along the z-axis. At the plane $z = 0$, it is considered to impinge perpendicularly on a material with flat, parallel faces containing emitting centers (see Fig. 1).

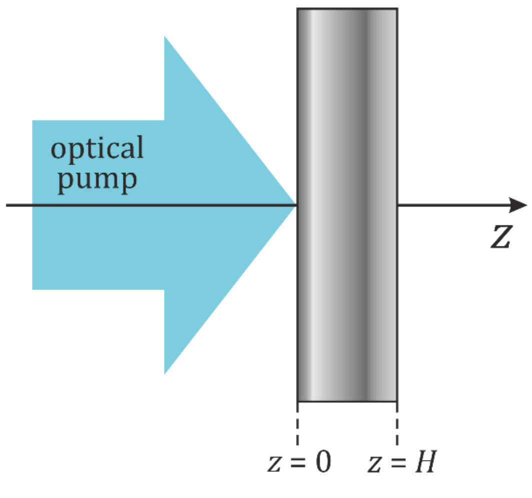

Figure 1. Optical excitation geometry. The material hosting the emitting centers has a thickness H. The density $N(z)$ of the emitting centers is represented by the intensity of the gray color in the figure and follows an arbitrary profile along the z-axis, while remaining uniform in the transverse plane Oxy, which is perpendicular to this axis.

The density of the emitting centers is assumed to be uniform in directions transverse to the z-axis, but varies along z according to an arbitrary function $N(z)$, which may not be known a priori. In order for these centers to emit Stokes-shifted photons, they must absorb photons from the pump; consequently, the pump intensity $P(z)$ decreases as it propagates through the material hosting the centers. To quantify this decrease, the differential equation governing the propagation of the intensity is

$$\frac{dP}{dz}(z) = -\alpha(z)P(z), \tag{1}$$

where $\alpha(z) \equiv \sigma N(z)$ is the material's absorption coefficient, and σ is the absorption cross section at the pump wavelength.

The solution to the above differential equation is

$$P(z) = P_0 \exp\left[-\int_0^z \alpha(z')\, dz'\right], \tag{2}$$

where $P_0 \equiv P(0)$ is the pump intensity at $z = 0$.[1]

2.2 Photoluminescence intensity

Under the assumption that any internal multiple reflection effects from the faces at $z = 0$ and $z = H$ of the solid can be neglected, and that the pump intensity is well below the saturation intensity [Sieg1986] of the emitting centers, which are considered as three- or four-level energy systems, the infinitesimal contribution to the PL intensity from a differential thickness dz located at depth z, with $0 \le z \le H$, is given by

$$dI(z) \propto N(z)P(z)dz \propto \alpha(z)P(z)dz, \tag{3}$$

so that the total PL intensity emitted by the whole solid is

$$I \propto P_0 \int_0^H \alpha(z) \exp\left[-\int_0^z \alpha(z')\, dz'\right] dz. \tag{4}$$

The analytical solution to this integral is[2]

$$I \propto P_0 \left\{1 - \exp\left[-\int_0^H \alpha(z)\, dz\right]\right\}. \tag{5}$$

By observing that the average value $\bar{\alpha}$ of the absorption coefficient over the interval $[0, H]$ is

$$\bar{\alpha} = \frac{1}{H}\int_0^H \alpha(z)\, dz, \tag{6}$$

Eq. (5) can be rewritten as

---

[1] Under the assumption that the real part of the solid's refractive index is spatially uniform, Eqs. (1) and (2) can be generalized to non-normal incidence by replacing $\alpha(z)$ with $\alpha(z)/\cos\theta$, where θ is the angle of wave propagation with respect to the z-axis within the solid.

[2] In the same case of non-normal incidence as described in the previous footnote, Eq. (5) can be generalized by replacing $\alpha(z)$ with $\alpha(z)/\cos\theta$ and P_0 with $P_0 \cos\theta$.

$$\frac{I}{P_0} \propto 1 - \mathrm{e}^{-\bar{\alpha} H}. \tag{7}$$

Since an analogous definition to Eq. (6) also applies to the average value $\bar{N}$ of the density of emitting centers, Eq. (7) can be rewritten as

$$\frac{I}{P_0} \propto 1 - \mathrm{e}^{-\sigma \bar{N} H}. \tag{8}$$

An interesting interpretation of Eqs. (7) and (8) is that the second term on the right-hand side—still neglecting internal multiple reflections—is equal to the total transmittance T of the solid at the pump wavelength, divided by the square of the single-face transmittance T_F between the solid and the external environment. Therefore, one can write[3]

$$\frac{I}{P_0} \propto 1 - \frac{T}{T_F^2}. \tag{9}$$

2.3 Correction of photoluminescence intensity

What would have been obtained if the gradual absorption of pump photons had not been taken into account? The answer can be found by omitting the variation of the pump intensity P along the z-axis, and instead assuming a constant value $P(z) \equiv P_0$. In this case, it can be easily verified that Eq. (4) would be trivially replaced by

$$I_0 \propto P_0 \int_0^H \alpha(z)\, dz\,, \tag{10}$$

where the symbol I_0 is used to distinguish this hypothetical case from the complete solution.

Recalling Eq. (6), one obtains

$$\frac{I_0}{P_0} \propto \bar{\alpha} H, \tag{11}$$

which corresponds to the first-order Taylor expansion with respect to $\bar{\alpha} H$ of the complete solution shown in Eq. (7). This observation provides a criterion to determine whether the absorption of the optical pump affects the PL emission: as intuition also suggests, such influence can be considered negligible when $\bar{\alpha} H \ll 1$. From a practical standpoint, using spectrophotometric measurements, the parameter $\bar{\alpha} H$ is approximately equal to 2.30 times the optical density of the solid, from which the optical density of an identical solid without emitting centers (commonly referred to as the 'blank' in laboratory terminology) has been subtracted—see Eqs. (7-9) and footnote 3. It should be noted that in both cases—whether pump absorption is considered or neglected—the PL intensity remains directly proportional to the pump intensity P_0. What differs between the two scenarios is the proportionality

[3] It can be verified that T/T_F^2 is equal to $10^{-(A-A_0)}$, which is approximately $\mathrm{e}^{-2.30\,(A-A_0)}$, where A and A_0 are the optical densities of the solid and of an identical solid not containing the absorbing-emitting centers, respectively.

factor, namely the slope of the line describing PL intensity as a function of P_0; in the presence of pump absorption, PL intensity increases more slowly with P_0.

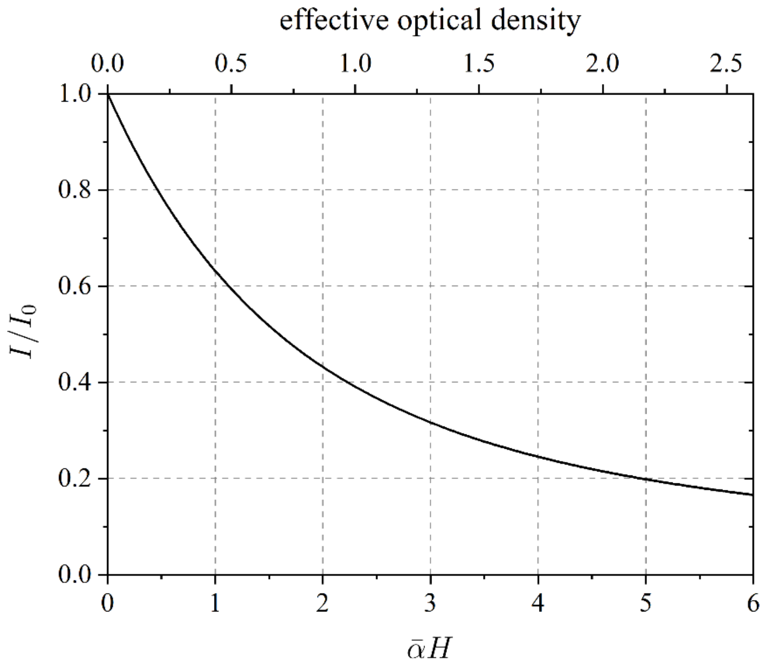

Figure 2. Ratio between the PL intensities calculated by considering and neglecting optical pump absorption, as a function of the parameter $\bar{\alpha}H$ (lower axis) and of the effective optical density (upper axis). This ratio corresponds to the reciprocal of the correction factor discussed in the text.

The impact of optical pump absorption as a function of the parameter $\bar{\alpha}H$ can be assessed by plotting the ratio $I/I_0 \equiv (1-\mathrm{e}^{-\bar{\alpha}H})/(\bar{\alpha}H)$ against this parameter—or equivalently, against the optical density A corrected by subtracting the optical density A_0 of the 'blank' sample, $A - A_0$ (see footnote 3), hereafter referred to as the 'effective optical density'. The resulting trend is shown in Fig. 2. To correct a PL measurement affected by pump absorption, the measured intensity I must be divided by the value of the ratio corresponding to $\bar{\alpha}H$ in the graph, yielding the corrected intensity

$$I_0 = \frac{\bar{\alpha}H}{1-\mathrm{e}^{-\bar{\alpha}H}} I = \frac{2.30(A-A_0)}{1-\mathrm{e}^{-2.30(A-A_0)}} I \,. \quad (12)$$

This procedure is the same of that reported by Skuja [Skuj2000] (Eq. (29), p. 85) for correcting PL excitation measurements, provided that: (a) the optical density referred to by the author is first corrected by subtracting that of the 'blank' sample, and (b) Skuja assumes the sample transmittance T to be equal to the absorption term $\mathrm{e}^{-\bar{\alpha}H}$, thereby implicitly considering the face transmittance T_F to be exactly equal to 1. As for the multiplicative factor 2.30 in the numerator, it is omitted by Skuja as it is non-essential, given that light intensity measurements are typically expressed in arbitrary units.

2.4. Creation of emitting centers by ionizing radiation

If the emitting centers are created through the interaction of the solid with ionizing radiation (e.g., color centers in LiF generated by X-ray or proton irradiation, etc.), it may be of interest to understand how, in the presence of optical pump absorption, the PL intensity varies as a function of the absorbed dose. Assuming the relationship between the center density N and the absorbed dose D follows the saturation-growth law initially introduced by Soshea et al. for the formation of color centers in MgO due to X-ray irradiation [Sosh1958], and later adopted for color centers in LiF [Picc2017, Mont2018, Picc2019, Nich2019a, Nich2020], one has

$$N = \widehat{N}\left(1 - \mathrm{e}^{-D/D_s}\right), \tag{13}$$

where $\widehat{N}$ is the maximum achievable density of centers in the material, and D_s is a parameter referred to in recent publications as the 'saturation dose'. The average density $\overline{N}$ across the thickness of the solid is then given by

$$\overline{N} = \frac{1}{H}\int_0^H N(z)\,dz = \widehat{N}\left[1 - \frac{1}{H}\int_0^H \mathrm{e}^{-D(z)/D_s}\,dz\right]. \tag{14}$$

In this expression, it is taken into account that the dose D may vary with depth z; its profile along the z-axis is essentially determined by the nature of the ionizing radiation, the composition of the solid material, and its thickness H.

By substituting Eq. (14) into Eq. (8), a composite function is obtained that is somewhat difficult to handle when attempting to establish a relationship between PL intensity and absorbed dose—for instance, to estimate the saturation dose via best-fit procedures. The main difficulty arises from the fact that the dependence on the absorbed dose is mathematically ambiguous in Eq. (14), precisely because the dose D, generally being depth-dependent, cannot be represented by a single-valued quantity. It is therefore necessary to identify a dependence on a more well-defined and stable quantity, such as the average dose $\overline{D}$ across the thickness H, which can intuitively serve this purpose and is calculated using an expression analogous to Eq. (6).

To enable this within a general formalism, it is convenient to define the auxiliary function

$$q(\mu) = \frac{D(\mu H)}{\overline{D}}, \quad \text{with } 0 \le \mu \le 1. \tag{15}$$

Observe that $q(\mu)$ is such that $q(\mu) \ge 0$, and

$$\int_0^1 q(\mu)\,d\mu = 1 \tag{16}$$

for any functional form of $D(z)$. The auxiliary function $q(\mu)$ introduced in this way is representative of the type of ionizing radiation and of the material, but not of the dose amount, as the latter can be quantified using the following relation

$$D(z) = \overline{D}q\left(\frac{z}{H}\right). \tag{17}$$

By substituting Eq. (17) into Eq. (14), one obtains

$$\overline{N} = \widehat{N}\left[1 - \frac{1}{H}\int_0^H \mathrm{e}^{-\overline{D}q(z/H)/D_s}\,dz\right] = \widehat{N}\left[1 - \int_0^1 \mathrm{e}^{-\overline{D}q(\mu)/D_s}\,d\mu\right]. \tag{18}$$

To obtain a result in a compact and self-consistent notation, the following additional auxiliary function is defined

$$\Omega(t) = \int_0^1 \mathrm{e}^{-q(\mu)t}\, d\mu\,, \tag{19}$$

so that the average density of centers can be expressed as

$$\bar{N} = \hat{N}\left[1 - \Omega\left(\frac{\bar{D}}{D_s}\right)\right]. \tag{20}$$

It should be noted that, in this general expression, the function Ω replaces the exponential function that appeared in Eq. (13). It can be shown that the function Ω satisfies the following properties

$$\Omega(0) = 1, \qquad \lim_{t\to\infty} \Omega(t) = 0, \qquad 0 < \Omega(t) \le 1. \tag{21}$$

In Eq. (21), the first property is trivial, the second holds provided that $q(\mu) > 0$ for all $\mu \in [0,1]$,[4] and the third can be easily demonstrated by observing that $0 < \mathrm{e}^{-q(\mu)t} \le 1$ for every $t \ge 0$.

By substituting Eq. (20) into Eq. (8), the following expression is finally obtained for the dependence of the PL intensity on the average absorbed dose in the solid

$$\frac{I}{P_0} \propto 1 - \mathrm{e}^{-\sigma\hat{N}H[1-\Omega(\bar{D}/D_S)]}\,. \tag{22}$$

The case in which pump absorption is completely neglected—that is, alternatively, when pump absorption has been corrected by applying Eq. (12)—is represented by Eq. (11), which, using the notation introduced above, becomes

$$\frac{I_0}{P_0} \propto \sigma\hat{N}H\left[1 - \Omega\left(\frac{\bar{D}}{D_s}\right)\right]. \tag{23}$$

As previously noted for Eq. (11), this expression corresponds to the first-order expansion of the exponential argument in Eq. (22).

2.5 Low-dose approximation

As can be easily verified, if the dose $D(z)$ is much smaller than D_s for every z in the interval $0 \le z \le H$, then the argument of the exponential function in the definition of $\Omega(t)$ (see Eq. (19)) satisfies $q(\mu)t \ll 1$ for every $\mu \in [0,1]$, given that $t \equiv \bar{D}/D_s$ when $\Omega(t)$ is used to compute the average density of centers, as shown in Eq. (20). Therefore, under this condition, the definition of $\Omega(t)$ can be approximated by

$$\Omega(t) \approx 1 - t\int_0^1 q(\mu)\, d\mu = 1 - t\,, \tag{24}$$

[4] It is worth stressing that the second property $\lim_{t\to\infty} \Omega(t) = 0$ is not satisfied when $q(\mu) = 0$ in some portions of the domain $\mu \in [0,1]$.

where the property stated in Eq. (16) has been used.

In this low-dose approximation, the average density of centers becomes directly proportional to the average dose. Indeed, by substituting Eq. (24) into Eq. (20), one obtains

$$\overline{N} \approx \beta\overline{D}\ , \tag{25}$$

where $\beta = \widehat{N}/D_s$. The parameter β can be identified with the center formation efficiency even for quantities averaged over the thickness H.[5] Thanks to the direct proportionality shown in Eq. (25), Eqs. (22) and (23) can be approximated at low doses by

$$\frac{I}{P_0} \propto 1 - \mathrm{e}^{-\sigma\beta H\overline{D}}, \tag{26}$$

$$\frac{I_0}{P_0} \propto \sigma\beta H\overline{D}. \tag{27}$$

From Eq. (26), it can be observed that, although the doses involved are much smaller than the saturation dose D_s, a nonlinear behavior still generally occurs, with an apparent saturation dose given by $\widetilde{D}_s = 1/(\sigma\beta H) = D_s/\big(\sigma\widehat{N}H\big)$. This nonlinear behavior is solely due to the absorption of the optical pump.

With regard to Eq. (27), which shows the PL intensity corrected for pump absorption, it can be normalized to the thickness H of the solid,

$$\frac{I_0}{P_0 H} \propto \sigma\beta\overline{D}. \tag{28}$$

By normalizing low-dose PL intensity measurements to the sample thicknesses, the slopes of the trends as a function of the average dose become mutually comparable even across samples of different thicknesses. This comparison is particularly useful, as the slope coefficients are proportional to the center formation efficiency β which, in principle, may vary from sample to sample depending on the irradiation conditions.

It should be noted that all considerations made for this low-dose case are independent of the specific form of the dose distribution $D(z)$.

3. Examples

In order to work with fixed numerical values in the following examples, let us focus on the representative case of visible-emitting F_2 and F_3^+ color centers in LiF, whose nearly overlapping absorption bands are peaked in the blue region of the spectrum at wavelengths of 444 and 448 nm, respectively [Mont2002]. Using Smakula's equation [Fowl1968], and assuming oscillator strengths in LiF of 0.28 for F_2 centers [Abuh1986] and 0.5 for F_3^+ centers,[6] inhomogeneous absorption-line broadening, a LiF refractive index of $n = 1.39$ at the center absorption wavelengths, and band widths

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[5] From Eq. (13), one can verify that the differential equation governing the center density as a function of dose is

$$dN/dD = \beta - \gamma N,$$

where $\beta \equiv \widehat{N}/D_s$ and $\gamma \equiv 1/D_s$ are the creation and annihilation coefficients of the centers, respectively. The creation coefficient can alternatively be interpreted as the center formation efficiency.

[6] The oscillator strength of F_3^+ centers in LiF is not available in the literature. Here, we arbitrarily set it to 0.5.

as specified in [Mont2002], the absorption cross section of both centers is estimated to be $\sigma \approx 2 \times 10^{-16}$ cm^{-2}. Assuming an average center density of $\bar{N} \approx 10^{17}$ cm^{-3} across the crystal thickness, and a typical LiF crystal thickness of $H = 1$ mm, it follows that $\sigma\bar{N}H \equiv \bar{\alpha}H = 2$ (optical density $A \approx 0.87$), a value high enough to significantly perturb the PL intensity measurement, as clearly visible in Fig. 2. If the average density were greater, $\bar{N} \approx 10^{18}$, an even more severe perturbation would be introduced due to $\sigma\bar{N}H \equiv \bar{\alpha}H = 20$ (optical density $A \approx 8.7$), that is, $I/I_0 \approx 0.05$.[7]

It is interesting to evaluate how measurements of PL intensity versus absorbed dose can be affected by optical pump absorption in the case of color centers in LiF crystals. In the following, some example scenarios are presented, which depend on the type of ionizing radiation used to generate the centers.

3.1 Uniform distribution

The first case examined is the trivial one of a homogeneous distribution of absorbed dose along the *z*-axis, as well as in the O*xy* plane: $D(z) \equiv \bar{D}$. In the case of color centers in LiF, such a distribution may result from irradiation with γ-rays, or X-rays with an attenuation length much greater than the crystal thickness H, or with protons of sufficiently high energy such that only the shallow (and practically flat) initial portion of the Bragg curve is contained within the thickness. In this case, it can be demonstrated that

$$q(\mu) = 1, \qquad \Omega(t) = \mathrm{e}^{-t}. \tag{29}$$

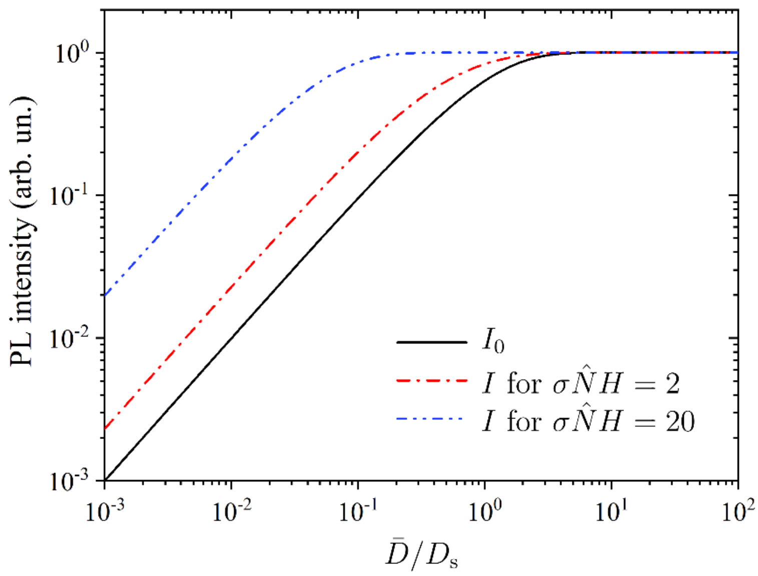

Figure 3. Theoretical PL intensity of color centers in a LiF crystal as a function of average dose, assuming uniform dose absorption. Comparison between the ideal case without pump absorption (I_0) and two representative cases where PL emission is affected by pump absorption. See the text for more details.

[7] These example values of maximum color center densities $\hat{N}$ in LiF are based on actual experimental findings regarding LiF thin films irradiated with γ-rays at doses up to 10^6 Gy [Nich2003].

Figure 3 shows the theoretical plots—computed using Eqs. (22), (23), and (29)—of the PL intensities[8] I and I_0 versus the average absorbed dose. In the figure, the PL intensities are normalized to their asymptotic values, while the average absorbed doses are normalized to the saturation dose. The two cases involving pump absorption are evaluated for the previously estimated parameter values $\sigma\hat{N}H = 2$ and $\sigma\hat{N}H = 20$. Note how, in both these cases, saturation of the PL emission starts at dose values significantly lower than that expected in the absence of pump absorption.

3.2 Exponential decay distribution

Another interesting case is that of a dose that decays exponentially along the thickness of the solid, $D(z) = D_0 \mathrm{e}^{-z/L}$, where $D_0 \equiv D(0)$ is the surface dose and L is the attenuation length in the material. Neglecting surface build-up effects, this could, for example, represent the dose deposited in a material by a beam of monochromatic X-rays. The thickness-averaged dose is

$$\overline{D} = \frac{1}{H}\int_0^H D_0 \mathrm{e}^{-z/L}\ dz = \frac{L}{H} D_0 \left(1 - \mathrm{e}^{-H/L}\right), \tag{30}$$

therefore the auxiliary function is

$$q(\mu) = \frac{H}{L}\frac{\mathrm{e}^{-\mu H/L}}{1 - \mathrm{e}^{-H/L}}. \tag{31}$$

The other auxiliary function $\Omega(t)$ admits the following analytical expression in terms of the special function Γ, known as the 'Euler incomplete gamma function',[9]

$$\Omega(t) = \frac{1}{H/L}\left\{\Gamma\left[0, \frac{(H/L)\mathrm{e}^{-H/L}\, t}{1 - \mathrm{e}^{-H/L}}\right] - \Gamma\left[0, \frac{(H/L)\, t}{1 - \mathrm{e}^{-H/L}}\right]\right\}. \tag{32}$$

The calculation of I/P_0 and I_0/P_0 using Eqs. (22) and (23) requires two parameters to be set: the first, as in the previous case of uniform dose distribution, is $\sigma\hat{N}H$; the second is the ratio H/L between the solid thickness and the X-ray attenuation length. As an example, let us consider monochromatic X-rays with an energy of 7 keV irradiating a LiF crystal of thickness $H = 1$ mm. According to the CXRO online calculator [CXRO], the penetration length of 7 keV X-rays in LiF is $L = 220$ μm, so that $H/L \approx 4.55$. As for the parameter $\sigma\hat{N}H$, let us again consider the two representative values of 2 and 20. Figure 4 shows the calculated dependencies of PL intensity on the mean absorbed dose, comparing the cases with and without optical pump absorption. In the figure, the PL intensities are normalized to their asymptotic values. Note that, similarly to the uniform dose case, pump absorption causes the curves to saturate at doses lower than that expected in the absence of saturation.

In general, it is observed that the shape of the absorption-less curve can dramatically change with the ratio H/L (see Fig. 5). A series of simulations varying this ratio in the range $0.001 \leq H/L \leq 100$ demonstrated that the effect of pump absorption causes a non-negligible departure from these ideal non-absorption curves when $\sigma\hat{N}H \gtrsim 1$.

[8] These PL intensities are meant to be those emitted by either F_2 or F_3^+ color centers. Possible PL quenching effects are neglected.

[9] The Euler incomplete gamma function is defined as $\Gamma(s, x) = \int_x^\infty t^{s-1}\mathrm{e}^{-t}\, dt$.

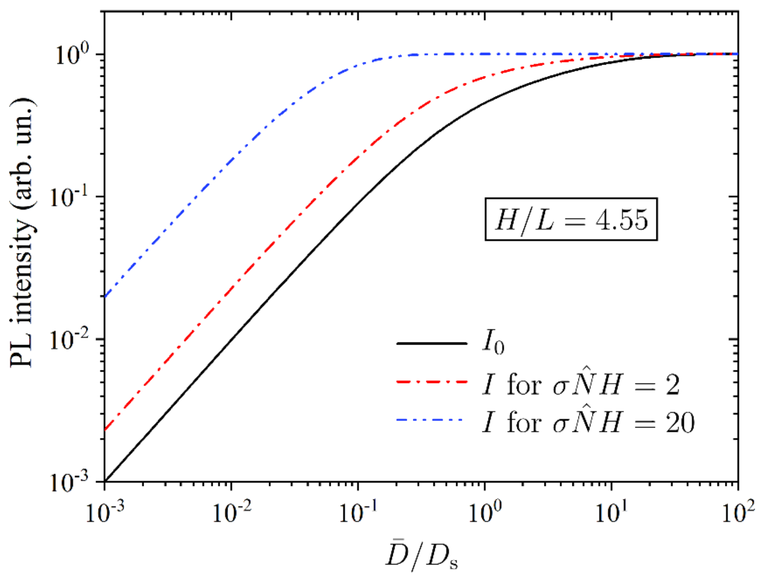

Figure 4. Theoretical PL intensity of color centers in a LiF crystal as a function of average dose, assuming exponential decay dose absorption. Comparison between the ideal case without pump absorption (I_0) and two representative cases where PL emission is affected by pump absorption. The assumed example ratio between crystal thickness and attenuation length is $H/L = 4.55$, corresponding to the absorption of 7 keV X-rays in LiF. See the text for more details.

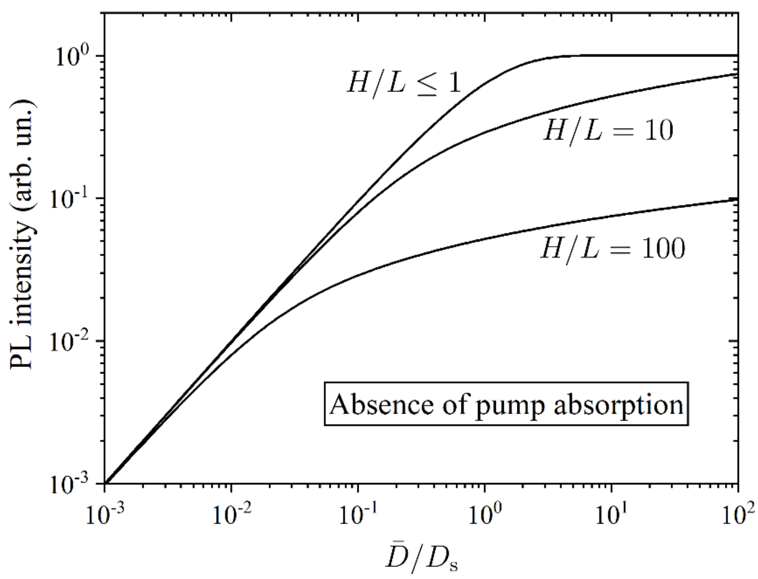

Figure 5. Theoretical PL intensity of color centers in a LiF crystal as a function of average dose, assuming exponential decay dose absorption and absence of pump absorption (I_0). The figure shows curves corresponding to different values of ratio H/L between crystal thickness and attenuation length of X-rays in LiF. The curves are normalized to their asymptotic values. See the text for more details.

3.3 Bragg-curve distribution

The final example concerns the irradiation of a LiF crystal slab with a beam of accelerated protons across its thickness. The dose deposition as a function of penetration depth by accelerated ions in materials follows the well-known Bragg curve. An approximate analytical expression to quantitatively evaluate proton Bragg curves in LiF, accurate for kinetic energies above approximately 10 MeV, was provided in [Nich2019b]. Although such an expression is available, no analytical form could be found for the auxiliary functions $q(\mu)$ and $\Omega(t)$ in this case. For this reason, the following calculations are carried out numerically.

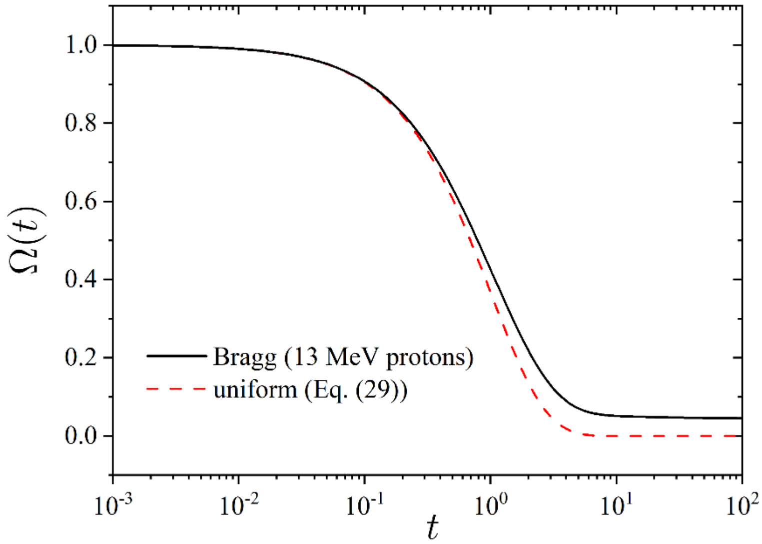

Figure 6. Numerically evaluated auxiliary function $\Omega(t)$ for the case of a Bragg-curve shaped distribution of absorbed dose induced across the thickness of a LiF crystal by 13 MeV protons. The exponential decay function corresponding to a uniformly distributed dose is also shown for comparison. See the text for more details.

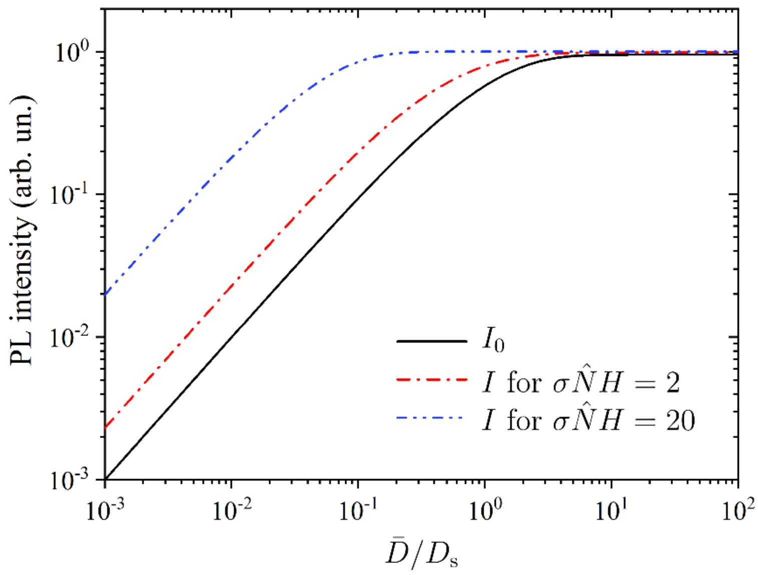

Figure 7. Theoretical PL intensity of color centers in a LiF crystal as a function of average dose, assuming Bragg-curve shaped dose absorption. Comparison between the ideal case without pump absorption (I_0) and two representative cases where PL emission is affected by pump absorption. The assumed energy of protons is 7 MeV. The curves are normalized to their asymptotic values. See the text for more details.

To analyze a specific case, the crystal slab thickness is assumed to be $H = 1$ mm, and the proton energy is set to 13 MeV, corresponding to a penetration range in LiF of approximately 930 μm, so that the Bragg curve spans almost entirely the crystal thickness. Figure 6 presents the numerically calculated auxiliary function $\Omega(t)$, along with a comparison to the exponential decay function corresponding to the uniform distribution case—see Eq. (29). Regarding the behavior of PL intensity as a function of dose, the cases $\sigma\hat{N}H = 2$ and $\sigma\hat{N}H = 20$ were considered here too and compared to the ideal case without pump absorption. The resulting curves, numerically calculated using Eqs. (22) and (23), are shown in Fig. 7. Note that the effect of pump absorption on these trends is comparable to that observed in the previous examples, resulting in an artificial reduction of the dose at which saturation occurs.

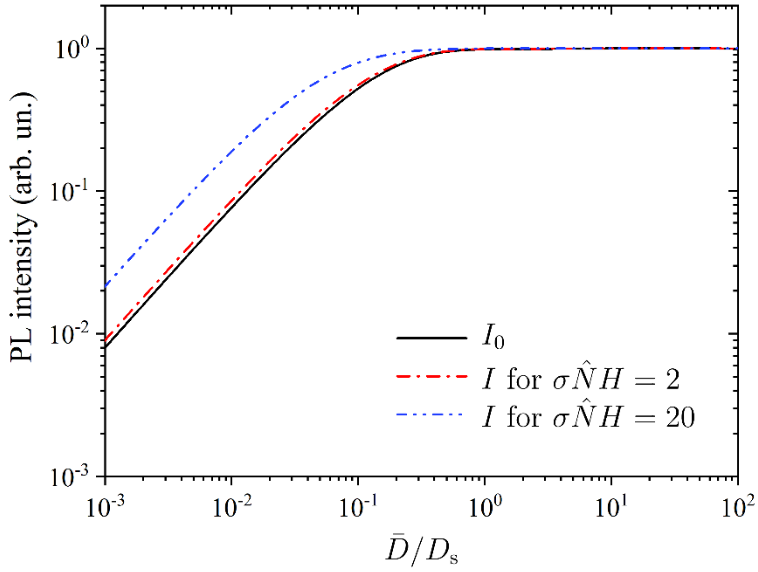

Figure 8. Theoretical PL intensity of color centers in a LiF crystal as a function of average dose, assuming Bragg-curve shaped dose absorption. Comparison between the ideal case without pump absorption (I_0) and two representative cases where PL emission is affected by pump absorption. The assumed energy of protons is 4 MeV. The curves are normalized to their asymptotic values. See the text for more details.

In the case of shallower proton penetration in LiF, the discrepancy between the saturation dose predicted by the ideal, absorption-free model and the apparent saturation dose accounting for pump absorption is reduced. The reason for this smaller difference is essentially ascribable to the fact that the optical pump is absorbed over a shorter depth and therefore undergoes less attenuation. This effect can be exemplified by considering, for instance, 4 MeV protons, whose penetration range R_0 in LiF is approximately 120 μm, as obtained using the Monte Carlo software SRIM [Zieg2010]. Numerical computations of PL intensity as a function of average dose, based on the depth-ionization profile simulated in SRIM, yielded the curves shown in Fig. 8.

An aspect that merits discussion—both in this specific case and, more broadly, in scenarios where the absorbed dose does not span the entire solid thickness—is the definition of average dose. Until now, the average absorbed dose $\bar{D}$ has been evaluated across the entire crystal thickness H. A more physically meaningful alternative may be to define the average dose over the effective penetration depth of the radiation depositing it. Let us label this alternative average dose as $\bar{d}$. For the present case, it can be readily shown that $\bar{d} = H\bar{D}/R_0$. Replotting the curves from Fig. 8 as functions of $\bar{d}/D_s$ instead of $\bar{D}/D_s$ yields the curves shown in Fig. 9. In this figure, the values of $\bar{d}/D_s$ at which the curves bend and saturate following the nearly linear growth phase show better agreement with the expected behavior ($\bar{d} \approx D_s$) compared to the curves in Fig. 8. This suggests that adopting the alternative definition $\bar{d}$ of average dose provides a more meaningful basis for plotting PL intensity as a function of dose, particularly when the plot is used to estimate the saturation dose value through curve fitting, provided that a preliminary correction of the measured PL intensity data is applied using the formula given in Eq. (12).

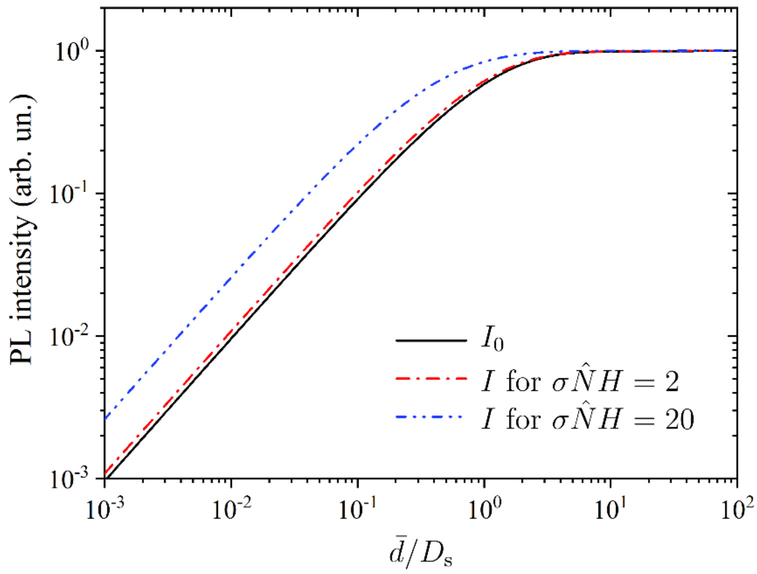

Figure 9. Theoretical PL intensity of color centers in a LiF crystal as a function of alternative average dose, assuming Bragg-curve shaped dose absorption. Comparison between the ideal case without pump absorption (I_0) and two representative cases where PL emission is affected by pump absorption. The assumed energy of protons is 4 MeV. The curves are normalized to their asymptotic values. See the text for more details.

4. Conclusions

When measuring PL intensity from a solid, it is essential to account for the gradual absorption of the optical pump as it traverses the material thickness, in order to quantitatively interpret the measurement results. In this Technical Report, a correction formula (Eq. (12)) has been derived, which is equivalent to the one reported by Skuja for correcting PL excitation measurements [Skuj2000].

The influence of pump absorption has been demonstrated through example cases involving PL emission from optically active color centers generated by ionizing radiation in LiF crystal slabs. Typically, the concentration of these centers increases almost linearly with the absorbed dose until saturation is reached. Consequently, a similar growth behavior is observed in the emitted PL intensity. By selecting appropriate representative values for the absorption cross section σ, crystal thickness H, and maximum center density $\hat{N}$, it has been demonstrated that pump absorption within the crystal can significantly reduce the dose value at which the PL intensity curve saturates.

The apparent saturation dose resulting from pump absorption is governed by Eq. (22), and for $\sigma\hat{N}H$ larger than ~2 is approximately equal to the actual saturation dose D_s divided by $\sigma\hat{N}H$, assuming the dose distribution spans the entire solid thickness.[10] This underestimation of D_s becomes less pronounced when the dose distribution covers a smaller thickness, as evident from the comparison between the example curves in Fig. 9 and those in Fig. 7. A limiting case occurs with thin films, where the reduced thickness allows the effects of pump absorption on saturation dose estimation to be considered negligible.

[10] It can be verified that a more precise estimation of the apparent saturation dose value, limited to the case of uniform dose distribution, is $-D_s \ln\left\{1 + \frac{1}{\sigma\hat{N}H} \ln\left[\frac{1}{\mathrm{e}} + \mathrm{e}^{-\sigma\hat{N}H}\left(1 - \frac{1}{\mathrm{e}}\right)\right]\right\}$. This is true if the value of $\bar{D}$ where the normalized function reaches a value of $1 - 1/\mathrm{e}$ is chosen as similarity criterion with the absorption-less curve.

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References

Abuh1986 L.H. Abu-Hassan, P.D. Townsend, *Ion implantation in LiF to form F and F_2 centres*, J. Phys. C: Solid State Phys. **19** (1986) 99-110.

CXRO CXRO online calculator: https://henke.lbl.gov/optical_constants/atten2.html

Fowl1968 W.B. Fowler, *Physics of Color Centers*, Academic Press (New York, NY), 1968.

Mont2002 R.M. Montereali, *Point defects in thin insulating films of lithium fluoride for optical microsystems, in: Handbook of Thin Film Materials*, ed. by H.S. Nalwa, Vol. 3, pages 399-431, Academic Press, (San Diego, CA), 2002.

Mont2018 R.M. Montereali, A. Ampollini, L. Picardi, C. Ronsivalle, F. Bonfigli, S. Libera, E. Nichelatti, M. Piccinini, M.A. Vincenti, *Visible photoluminescence of aggregate colour centres in lithium fluoride thin films for low-energy proton beam radiation detectors at high doses*, J. Lumin. **200** (2018) 30–34.

Nich2003 E. Nichelatti, R.M. Montereali, M. Montecchi, T. Marolo, *Optical properties of rough, inhomogeneous lithium fluoride films with colour centres*, J. Non-Cryst. Solids **322** (2003) 117-121.

Nich2019a E. Nichelatti, M. Piccinini, A. Ampollini, L. Picardi, C. Ronsivalle, F. Bonfigli, M.A. Vincenti, R.M. Montereali, *Modelling of photoluminescence from F_2 and F_3^+ colour centres in lithium fluoride irradiated at high doses by low-energy proton beams*, Opt. Mater. **89** (2019) 414–418.

Nich2019b E. Nichelatti, C. Ronsivalle, M. Piccinini, L. Picardi, R.M. Montereali, *An analytical approximation of proton Bragg curves in lithium fluoride for beam energy distribution analysis*, Nucl. Instrum. Methods Phys. Res. B **446** (2019) 29-36.

Nich2020 E. Nichelatti, M. Piccinini, C. Ronsivalle, L. Picardi, M. Vincenti, R. Montereali, *Evaluation of saturation dose in spatial distributions of color centers generated by 18 MeV proton beams in lithium fluoride*, Nucl. Instrum. Methods Phys. Res. B **464** (2020) 100–105.

Picc2017 M. Piccinini, E. Nichelatti, A. Ampollini, L. Picardi, C. Ronsivalle, F. Bonfigli, S. Libera, M.A. Vincenti, R.M. Montereali, *Proton beam dose mapping via color centers in LiF thin film detectors by fluorescence microscopy*, EPL **117** (2017) 37004.

Picc2019 M. Piccinini, E. Nichelatti, C. Ronsivalle, A. Ampollini, G. Bazzano, F. Bonfigli, P. Nenzi, V. Surrenti, E. Trinca, M. Vadrucci, M.A. Vincenti, L. Picardi, R.M. Montereali, *Visible photoluminescence of color centers in LiF crystals for advanced diagnostics of 18 and 27 MeV proton beams*, Radiat. Meas. **124** (2019) 59–62.

Sieg1986 A.E. Siegman, *Lasers*, University Science Books (Mill Valley, CA), 1986.

Skuj2000 L. Skuja, *Optical properties of defects in silica, in: Defects in SiO_2 and Related Dielectrics: Science and Technology*, ed. by G. Pacchioni, L. Skuja, and D.L. Griscom, NATO Science Series – II. Mathematics, Physics and Chemistry, Vol. 2, pages 73-116, Kluwer Academic Publishers (The Netherlands), 2000.

Sosh1958 R.W. Soshea, A.J. Dekker, J.P. Sturtz, *X-ray induced color centers in MgO*, J. Phys. Chem. Solids **5** (1958) 23-33.

Zieg2010 J.F. Ziegler, M.D. Ziegler, J.P. Biersack, *SRIM – The stopping and range of ions in matter*, Nucl. Instrum. Methods Phys. Res. B **268** (2010) 1818-1823.

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