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## Energy resolution limits of amplified scintillators

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### ABSTRACT

In this paper, we analyze the potential improvement in the statistical limit to the energy resolution of a scintillator using a scintillator amplification technique. The work excludes potentially complicating factors, such as non-linearity in light yield, since these factors are dependent upon the particular choice of detector materials. The results show that energy resolutions typical of semiconductors are technically feasible for scintillators. In the example presented, introducing scintillation multiplication improved the energy resolution limit from a few percent to below 1%.

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## 1. Introduction

We recently proposed [1] a method to overcome an inherent problem associated with scintillator detectors. In all inorganic scintillator materials in use today, each electron–hole pair, formed by the ionization process, yields only a fraction of a scintillator photon. New materials are being researched that optimize this fraction. These photons must then flow into an exit window and be converted to an electron, a process that can be rather inefficient. Under the best of conditions, the energy resolution of the scintillator will be statistically limited by the number of carriers at their lowest population, e.g., after the photocathode or photodiode. But if each electron–hole pair led to multiple scintillation photons, then it might be possible to match the energy resolution of semiconductor detectors, and open new application areas as well.

One approach to breaking the limit of one photon per electron is to choose an activator atom in a host material that will undergo a change that can be repeatedly sampled an arbitrary number of times and then “reset”. The processes involved must be fast enough to accommodate a desired radiation interaction rate.

An example of this general concept is depicted in Fig. 1. A divalent rare earth ion ( $\text{RE}^{2+}$ ) is produced as the result of an electron having been captured on a trivalent activator atom ( $\text{RE}^{3+}$ ). The scintillator is always illuminated by laser photons that excite any ( $\text{RE}^{2+}$ ), which then quickly de-excites by a typical

fluorescence cascade that generates light at one or more wavelengths such as  $\lambda_1$ ,  $\lambda_2$  or  $\lambda_3$  in the figure. As long as  $\text{RE}^{2+}$  persists, by cyclically stimulating  $\text{RE}^{2+}$  to an excited state, and measuring either  $\lambda_1$ ,  $\lambda_2$  or  $\lambda_3$ , one can obtain as many signal photons as desired by adjusting the observation time. With a single pulse of suitably short wavelength, the  $\text{RE}^{2+}$  atoms may then be converted back to  $\text{RE}^{3+}$  to reset the detector. For any chosen accumulation time, the signal output from any voxel of the detector is proportional to the number of valence transitions induced by the incident radiation since the last reset.

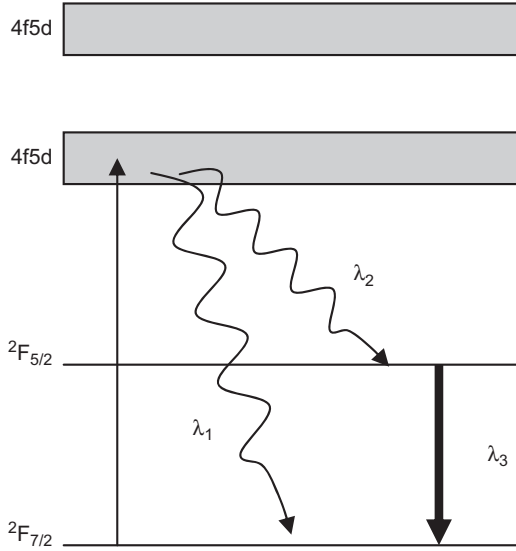
If scintillation yield is removed as a limiting factor, this could enable different materials to be candidates for detectors. In this paper, we deliberately avoid discussions of particular detector materials or physical instantiations of the idea, but instead model how this approach would impact the theoretical energy resolution of such detectors, and in so doing, identify the parameter that will likely prove to be a key factor when looking for new materials. Our approach will be to examine each step in the pulse formation process, beginning with the birth of the electron–hole pairs.

## 2. Formation of the initial electron–hole pairs

Suppose the incident gamma ray deposits an energy  $E_{\text{dep}}$  and produces  $n_{\text{eh}}$  electron–hole pairs. Since this is a random process, we expect  $n_{\text{eh}}$  to have a mean value of  $\bar{n}_{\text{eh}}$  and variance  $\sigma_{\text{eh}}^2 = F\bar{n}_{\text{eh}}$ , where  $F$  is the Fano factor to account for non-Poisson electron–hole pair production in the material. As a general rule,  $n_{\text{eh}} = E_{\text{dep}}/3E_{\text{gap}}$  where  $E_{\text{gap}}$  is the energy gap of the detector material [2].

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**Fig. 1.** Energy level diagram of a divalent rare earth ion ( $RE^{2+}$ ) produced by capture of an electron by the corresponding trivalent ion ( $RE^{3+}$ ). A cycle of optical excitation followed by a cascade of luminescence on three wavelengths is shown.

### 3. Valence change

Once formed, the electrons and the holes will migrate to an activator atom. We shall assume that the electron and the hole do not arrive simultaneously, but rather there is a time period in which the valence state of the activator is altered. Define  $n^+$  to be the number of valence-modified activator atoms, where we expect

$$\bar{n}^+ = \bar{n}_{eh}\varepsilon_A$$

where  $\varepsilon_A$  is the efficiency by which electron–hole pairs produce valence-modified activator atoms.

Using  $\sigma^2_x = \bar{x}^2 - \bar{x}^2$  and closely paralleling the approach of Ref. [3], we write the expression for the variance in  $\bar{n}^+$  as

$$\sigma_{n^+}^2 = \left( \sum_{n_{eh}} \sum_{n^+} n_+^2 \Pr(n^+ | n_{eh}) \Pr(n_{eh} | \bar{n}_{eh}) \right) - (\bar{n}_{eh}\varepsilon_A)^2$$

where  $\Pr$  is a conditional probability. We will assume  $\Pr(n^+ | n_{eh})$  follows a Binomial distribution, but make no assumption regarding the  $\Pr(n_{eh} | \bar{n}_{eh})$  distribution.

Then performing the sum first over  $n^+$  yields

$$\sigma_{n^+}^2 = \sum_{n_{eh}} [\varepsilon_A n_{eh} (1 - \varepsilon_A) + (\varepsilon_A n_{eh})^2] \Pr(n_{eh} | \bar{n}_{eh}) - (\bar{n}_{eh}\varepsilon_A)^2$$

and after a bit of algebra, one ends up with

$$\frac{\sigma_{n^+}^2}{\bar{n}_+^2} = \frac{1}{n_{eh}} \left[ \left( \frac{1 - \varepsilon_A}{\varepsilon_A} \right) + F \right] \quad (1)$$

### 4. Photon multiplication

Once a valence change has occurred, the resulting atom has optical properties that differ from those of the unmodified atom. By repeatedly exciting this valence-changed atom to one of its unique excited optical states, and observing its emissions, one can produce  $m$  scintillation photons per atom. The value of  $m$  is governed in practice by the ratio of excitation and emission frequencies, and the total observation time. From any single atom with a probability/time of an emission from an excited state given by  $\lambda$ , we expect that the variance in the number of photons

emitted during an observation time  $T$  will be Poisson, i.e.,

$$\sigma_m^2 = (\lambda T) = \bar{m} \quad \text{or} \quad \frac{\sigma_m^2}{\bar{m}^2} = \frac{1}{\bar{m}} = \frac{1}{\lambda T} = \frac{\tau}{T}$$

where  $\tau = 1/\lambda$ . If  $\tau = 1$  ns, and  $T = 1$   $\mu$ s, then  $m = 10^3$ . We shall see later that significantly smaller values of  $m$  are likely to be sufficient. For  $n^+$  atoms, the variance of the multiplication for the ensemble will be the variance in  $m$  divided by  $n^+$ , so

$$\frac{\sigma_M^2}{\bar{M}^2} = \frac{1}{n^+ \bar{m}} = \frac{\tau}{n_{eh}\varepsilon_A T} \quad (2)$$

### 5. Photon collection and photoelectron production

After the scintillation photons are created, they must then reach the photo-readout (governed by the light collection efficiency) and create photoelectrons (limited by the quantum efficiency). We will follow the convention of lumping these together as a photon conversion efficiency,  $\eta$ . If we assume that we have  $N$  photons, then the variance in the number of photoelectrons  $n_{pe}$  that are generated will be

$$\sigma_{pe}^2 = \overline{n_{pe}^2} - \bar{n}_{pe}^2 = \sum_{N=0}^{\infty} \sum_{n_{pe}=0}^N n_{pe}^2 \Pr(n_{pe} | N) \Pr(N | \bar{N}) - \bar{n}_{pe}^2$$

As before, we assume  $\Pr(n_{pe} | N)$  follows a Binomial distribution and so it will have a variance of  $\eta N(1 - \eta)$ . Recognizing that

$$\sum_{n_{pe}=0}^N n_{pe}^2 \Pr(n_{pe} | N) = \overline{n_{pe}^2} = \sigma_{pe}^2 + \bar{n}_{pe}^2 = \eta(1 - \eta)N + (\eta N)^2$$

and then substituting into the previous expression yields

$$\sigma_{pe}^2 = \left( \sum_{N=0}^{\infty} (\eta(1 - \eta)N + (\eta N)^2) \Pr(N | \bar{N}) \right) - \bar{n}_{pe}^2$$

which, after applying the same technique to the sum over  $N$ , yields

$$\sigma_{pe}^2 = \eta(1 - \eta)\bar{N} + \eta^2 \sigma_N^2 \quad \text{or} \quad \frac{\sigma_{pe}^2}{n_{pe}^2} = \left( \frac{1 - \eta}{\eta} \right) \frac{1}{\bar{N}} + \frac{\sigma_N^2}{\bar{N}^2} \quad (3)$$

Note that we have made no assumption about the underlying distribution for  $N$ .

### 6. The statistical limit to the energy resolution

If we recall that  $N = n^+ M$ , then using

$$\frac{\sigma_N^2}{\bar{N}^2} = \frac{\sigma_M^2}{\bar{M}^2} + \frac{\sigma_{n^+}^2}{\bar{n}_+^2}$$

and substituting in values for the relative variances from above, we arrive at the result we seek:

$$\frac{\sigma_{pe}^2}{n_{pe}^2} = \frac{1}{n_{eh}} \left( F + \frac{1}{\eta \varepsilon_A} \left( \frac{\tau}{T} \right) + \left( \frac{1 - \varepsilon_A}{\varepsilon_A} \right) \right) \quad (4)$$

The first term in Eq. (4) is the same as that used to predict the energy resolution of semiconductor detectors [2]. The second term accounts for the degradation in energy resolution due to the light collection and quantum inefficiency. Low values for these can be overcome using the scintillator amplification process [1]. Since small values of  $\tau/T$  imply better resolution, faster scintillators or longer observation times are desirable. Assuming other effects, such as scintillator non-linearity, are not problematic, the third term presents the materials challenge to achieving the best energy resolution using this new approach. We hence must seek detector materials for which  $\varepsilon_A \rightarrow 1$ , i.e., materials having an efficient valence-change process. We optimistically note that conventional

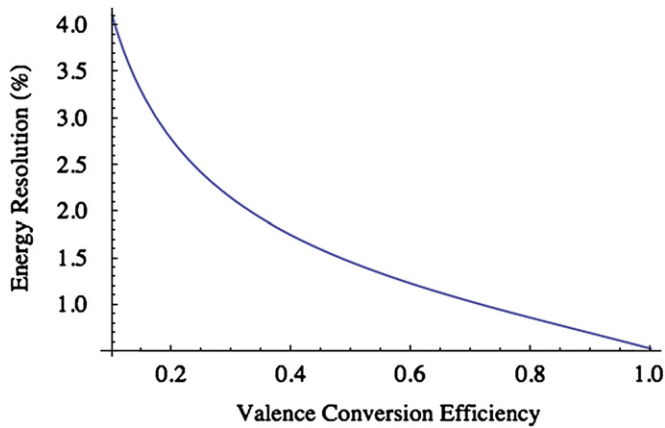


Fig. 2. Energy resolution as a function of  $\varepsilon_A$ , for  $m=1000$  and typical scintillator values listed in the text.

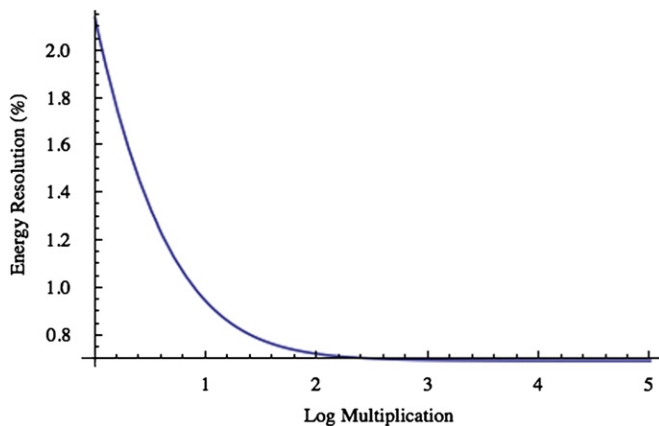


Fig. 3. Energy resolution as a function of  $\text{Log}[\text{Multiplication}]$  for  $\varepsilon_A=0.9$  and typical scintillator values listed in the text.

scintillators have proven to be efficient at converting electron–hole pairs into scintillation photons.

Using this expression with typical values for a conventional scintillator, i.e., choosing  $\eta = 0.5$ ,  $\varepsilon_A = 0.9$ ,  $F = 0.15$ , and assuming 30,000 electron–hole pairs from a 662 keV interaction, one obtains an energy resolution of  $\sim 2\%$ , which evidence shows to be about the limit of conventional scintillator technology. For comparison, when including a multiplication factor of  $m=1000$ , the theoretical energy resolution limit improves dramatically to 0.7% as shown in Fig. 2. Interestingly, Fig. 3 shows that lower values of multiplication, perhaps only 100, would be sufficient to achieve most of the expected gain in energy resolution. Finally, Fig. 4 shows

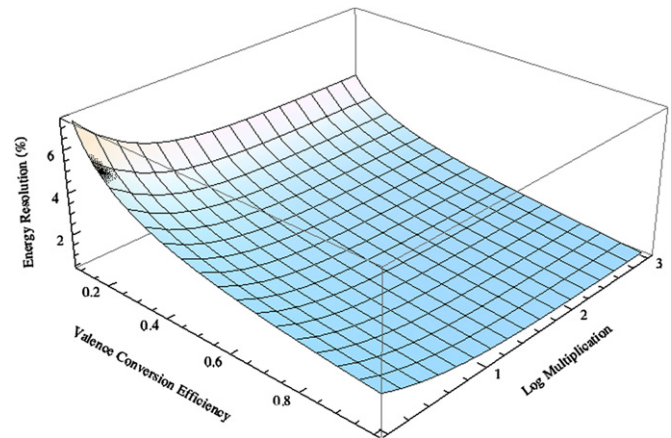


Fig. 4. Energy resolution as a function of  $\varepsilon_A$  and multiplication, using typical scintillator values listed in the text.

the statistical limit to scintillator energy resolution as a function of both  $\varepsilon_A$  and the logarithm of the multiplication. This figure shows the tradeoff of valence conversion efficiency with multiplication to achieve a given energy resolution.

## 7. Summary

In this paper, we quantified the improvement in the statistical limit to the energy resolution of a scintillator using the scintillator amplification technique we first presented in Ref. [1]. We have not addressed potentially complicating factors, such as non-linearity in light yield, since these factors are dependent upon the choice of detector materials. The results of this work have shown that energy resolutions typical of semiconductors should be feasible for scintillators. In the example presented, introducing scintillation multiplication improved the energy resolution limit to below 1%. The next step is to explore detector materials, beyond those demonstrated in Ref. [1] that are suitable candidates to validate this concept.

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