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S. E Letant, T.-F. Wang

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S. E. Létant [a)] and T.-F. Wang

Chemistry and Materials Science Directorate, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, California, 94550

We recently demonstrated the ability of semiconductor quantum dots to convert alpha radiation into visible photons. In this letter, we report on the scintillation of quantum dots under gamma-ray irradiation, and compare the energy resolution of the 59 keV line of Americium 241 obtained with our quantum dot-glass nanocomposite material to that of a standard sodium iodide scintillator. A factor 2 improvement is demonstrated experimentally and interpreted theoretically using a combination of energy-loss and photon transport models. These results demonstrate the potential of quantum dots for room-temperature gamma-ray detection, which has applications in medical imaging, environmental monitoring, as well as security and defense.

[a)] Author to whom correspondence should be addressed; electronic mail: *letant1@llnl.gov*

Present technology in gamma radiation detection suffers from flexibility and scalability issues. For example, bulk Germanium provides fine energy resolution (0.2 % energy resolution at 1.33 MeV)[1-3] but requires operation at liquid nitrogen temperature. On the other hand, Cadmium-Zinc-Telluride is a good room temperature detector (1 % at 662 keV) but the size of the crystals that can be grown is limited to a few centimeters in each direction.[4] Finally, the most commonly used scintillator, Sodium Iodide (NaI), can be grown as large crystals but suffers from a lack of energy resolution (7 % energy resolution at 662 keV).[5]

Recent advancements in nanotechnology[6-10] have provided the possibility of controlling materials synthesis at the molecular level. Both morphology and chemical composition can now be manipulated, leading to radically new material properties due to a combination of quantum confinement and surface to volume ratio effects. One of the main consequences of reducing the size of semiconductors down to nanometer dimensions is to increase the energy band gap, leading to visible luminescence, which suggests that these materials could be used as scintillators. The visible band gap of quantum dots would also ensure both efficient photon counting (better coupling with photomultipliers optimized for the visible region), and high photon output (smaller individual photon energy results in more photons produced) at room temperature, which is essential for effective Poisson counting (the energy resolution $\Delta E/E$ is inversely proportional to the square root of the number of photons collected).

We recently demonstrated the ability of semiconductor quantum dots to convert alpha radiation into visible photons, and interpreted the results using the MCNP (Monte Carlo for N-particles) model.[11] In this letter, we report on the scintillation of quantum dots

under gamma irradiation, and compare the energy resolution of the 59 keV line of Americium 241 obtained with our nanocomposite material to that of a standard NaI scintillator. The data is interpreted using a combination of energy-loss and photon transport models, which allows to estimate the number of photons produced in the material at a given energy.

Porous VYCOR® was purchased from Advanced Glass and Ceramics (Holden, MA) in 1/16 inch thick sheets. As received, the material is constituted of an array of interconnected pores with a diameter of 4 nm and is opalescent. The porous glass matrix was slowly dissolved for 4 days in an aqueous solution containing 1 % of hydrofluoric acid and 20 % of ethanol per volume, rinsed in ethanol, and dried in air. The purpose of this step was to slightly enhance the pore size to 10 nm and to obtain a clear matrix.[11] CdSe/ZnS core shell quantum dots with a luminescence output at 510 nm (well matched with the response curve of our photo-detector) were purchased from Evident Technologies (Troy, NY) and suspended in toluene at a concentration of 10 mg / mL. The dry 'thirsty' porous glass pieces were immersed in the solutions of dots for 48 H with continuous stirring in order to allow homogeneous diffusion of the dots into the nano-porous host matrix. They were then air-dried in order to evaporate the solvent.

Absorbance spectra of the samples were recorded using a UV-vis spectrometer (Cary 100, Varian Inc.) and emission spectra were recorded using a fluorimeter fitted with an optic fiber (Cary Eclipse, Varian Inc.). This data (not shown) was inputted into the simulations.

An alpha source ($^{243\text{-}244}$Cm, 0.2 μCi) was placed in contact with one side of the 1/16 inch thick porous glass sample and a PMT (model R1924A from Hamamatsu, 15%

quantum efficiency at 510 nm) probing a 1.5 cm diameter area rested directly on the other side to count visible photons coming out of the material. Photons coming out of the nano-composite samples under alpha irradiation were integrated for 10 H with an amplifier and a multi-channel analyzer. Figure 1a shows the scintillation output recorded through the sample with the $^{243\text{-}244}$Cm source and corrected from background radiation, demonstrating conversion of the alpha radiation into visible photons.

In order to asses the scintillation of the material under gamma irradiation, the alpha source was then replaced by an Americium point source (^{241}Am, 1 μCi). Since the gamma-ray stopping power of a material increases exponentially with its thickness, density, and attenuation coefficient, and increases with the gamma-ray energy, recording scintillation on the un-optimized porous glass-quantum dot composites requires both a long integration time (in the order of days) and a low energy source (below 100 keV). The scintillation output of our nanocomposite material was recorded for 3 days and corrected from background radiation (see Figure 1b). A distinctive peak was observed in the presence of the Americium source that we attribute to the 59 keV line of ^{241}Am. ^{241}Am predominantly decays (82.5 %) to the second excited state of ^{237}Np and then emits a 59.54 keV gamma-ray to the ground state of ^{237}Np. The Gaussian fit of the peak shown in inset of Figure 1b provided a value of the energy resolution ΔE/E of 15 % at 59 keV. The scintillation of a standard 1'' x 1'' NaI scintillator crystal was recorded in the same conditions as a reference (see Figure 2), and the Gaussian fit of the experimental curve provided a value of the energy resolution ΔE/E of 30 % at 59 keV, which is in agreement with the literature.[12] This data indicates a factor 2 improvement in energy resolution of our un-optimized nanocomposite material over a standard NaI crystal.

In order to interpret these experimental results in terms of number of photons generated in the material at a given energy, we performed simulations including one million interactions, using a combination of MCNP and DETECT 2000 programs in history mode.[13,14] At this low gamma-ray energy, the interactions are mostly terminated by the photo-absorption mechanism. While it is difficult to interpret scintillation data recorded with an alpha source due to its strong dependence on surface effects and local quantum dot density,[11] low energy gamma-ray interactions, which are dominated by local photo-absorption, can provide reliable information on the number of photons generated via photo-emission of the quantum dots. The interaction history combined with light transport simulated according to the specific properties of the scintillator medium (such as the wavelength-dependent absorption and emission coefficients of the emitters, the nature of the matrix (index of refraction, scattering length, absorption length), the scintillation decay times, and the quantum efficiency of the photomultiplier) allow to identify losses in the material, and to estimate the number of photons produced by our semiconductor quantum dot scintillators.

For an ideal scintillator, the energy resolution R, is given by: $R = (\Delta E/E)_{fwhm} = 2.35 \cdot ((1+v(M))/(N \cdot p))^{1/2}$, where N is the average number of photons generated at a given energy E, v(M) is the variance in the multiplication factor of the PMT (for a typical 10 stages PMT[15] with a gain of 2.10^6, v(M) is approximately 0.08), and p is the average transport efficiency.[15] $\Delta E/E$ is measured experimentally, v(M) is a known constant, and combined histories and transport simulations can provide an estimate of p, the average transport efficiency of the material. The only unknown parameter is then N, the number of photons generated in the material under an incident gamma energy E.

The value of the average transport efficiency p, obtained from the MCNP-DETECT 2000 simulation is 0.063 ± 0.002. By inserting p in the energy resolution the formula discussed above, the average photon output of our material under 59 keV gamma-ray irradiation is estimated to be close to 4,210 (while the number of photons generated by an NaI crystal under the same conditions is only 2,600). For higher energies the projected light output would be about 70,600 photons at 1 MeV, assuming the quantum dot medium has a linear response, which is a factor 1.75 better than the NaI crystal for which N is typically 40,000.

We reported a factor 2 improvement in energy resolution of an un-optimized nanoporous glass-quantum dot composite material over a standard NaI crystal, using the 59 keV line of an Americium gamma source. The data was interpreted theoretically in terms of number of photons generated in the material under gamma irradiation. Although we demonstrated that semiconductor quantum dot have an adequate photon output at low gamma energy, scintillation and linearity will have to be studied for higher gamma-ray energies (i.e., depleted Uranium line at 1001 keV). To produce the large quantum dot scintillators required for these studies, we will need to use densely packed high-Z semiconductor quantum dots. The linearity of the scintillation output will then be investigated in terms of number of photons produced in the scintillator versus incident gamma-ray energy.

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FIGURE CAPTIONS

FIG. 1. Scintillation output of a 25 mm thick quantum dot-nanoporous glass composite under a) alpha irradiation with a Curium 243-244 source, and b) gamma irradiation with an Americium 241 source. Both spectra were corrected from background radiation. A Gaussian fit of the 59 keV line of Americium 241 shown in inset indicates an experimental energy resolution $\Delta E/E$ of 15 % at this energy.

FIG. 2. 59 keV line of an Americium 241 source recorded with a standard 1" x 1" NaI scintillator crystal and corrected from background radiation (dotted line). The corresponding Gaussian fit (solid line) indicates an experimental energy resolution $\Delta E/E$ of 30 % at this energy.

Figure 1

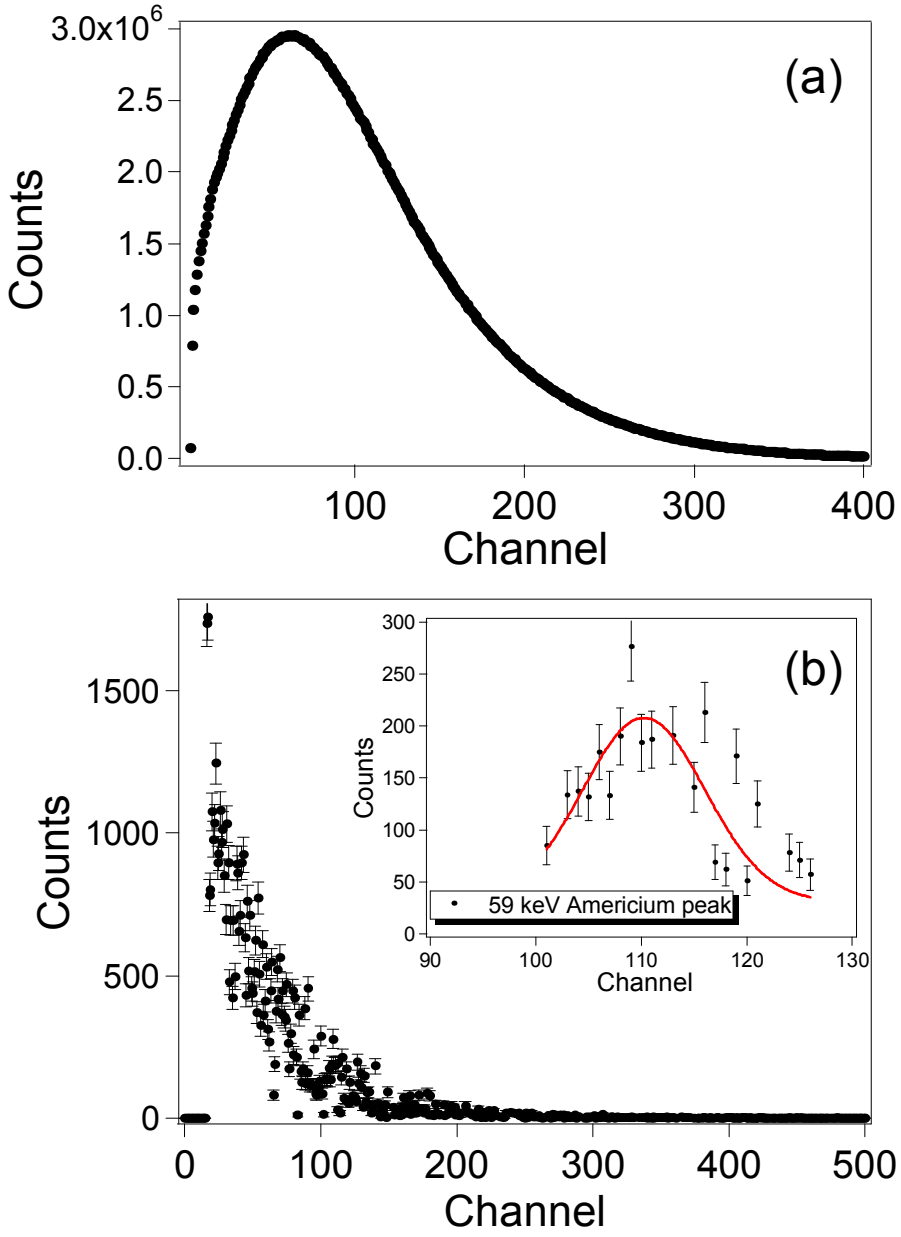
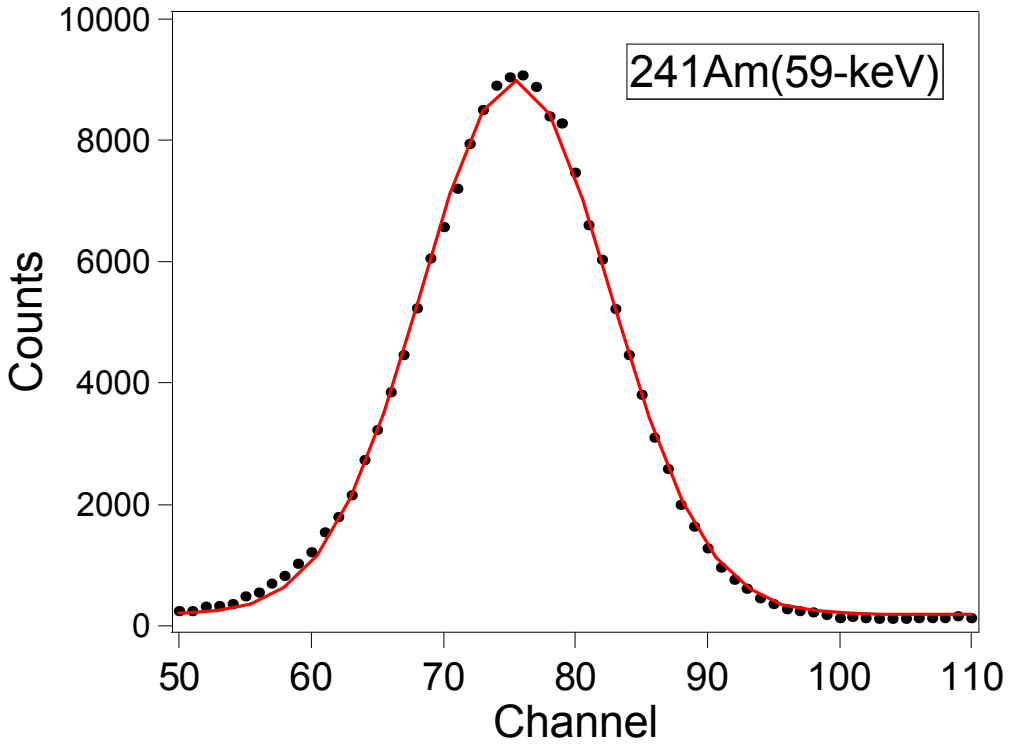

3.0x10^6
2.5
2.0
1.5
1.0
0.5
0.0
Counts
(a)
100
200
300
400
Channel
1500
1000
500
0
Counts
(b)
300
250
200
150
100
50
0
Counts
59 keV Americium peak
90
100
110
120
130
Channel
0
100
200
300
400
500
Channel

Figure 2


241Am(59-keV)
Counts
Channel
10000
8000
6000
4000
2000
0
50
60
70
80
90
100
110