Energy calibration of plastic scintillator by pair production of 4.44 MeV γ's

Melisa Ozen[1,2], John A. Behr[1,3,*] Michelle Khoo[3], Felix Klose[3], Alexandre Gorelov[1], and Dan Melconian[4,5]
[1] *TRIUMF. 4004 Wesbrook Mall, Vancouver, BC V6T 2A3, Canada*
[2] *Department of Physics, University of Ottawa, Ottawa, ON K1N 6N5, Canada*
[3] *Department of Physics and Astronomy, University of British Columbia, Vancouver, BC V6T 1Z1, Canada*
[4] *Cyclotron Institute, Texas A&M University, 3366 TAMU, College Station, Texas 77843-3366, USA and*
[5] *Department of Physics and Astronomy, Texas A&M University, 4242 TAMU, College Station, Texas 77843-4242, USA*

We demonstrate energy calibration of a 40x88 mm plastic scintillator at the relatively high energy of 3.42 MeV, observing the double-escape peak from pair production of 4.44 MeV γ-rays by tagging 511 keV annihilation radiation in two high-Z scintillators. The source is a standard commercial neutron source using ^{241}Am encapsulated with ^{9}Be, which has a reaction branch feeding the first $I^{\pi}=2^{+}$ state of ^{12}C. We also measure the energy resolution. By comparison with conventional lower-energy sources, the precision allows us to find a systematic difference between calibrations using γ-rays compared to a conversion electron source, likely produced by slightly nonuniform light collection. The energy resolution improves with photon statistics, yet shows a component constant with energy. The experimental lineshape response tail exceeds a GEANT4 simulation of bremsstrahlung escape and energy loss from the detector edges, which excess we can qualitatively explain from observed backgrounds, likely involving neutrons from the source.

I. INTRODUCTION

We are using relatively large plastic scintillators for precision measurements, requiring β energy spectroscopy with endpoints near 5 MeV [1]. We have recently changed scintillator readout from photomultiplier tube to silicon photomultiplier technology, to minimize gain changes from the switching magnetic fields of our magneto-optical trap, creating a need to test linearity of gain. The internal conversion electrons from ^{207}Bi decay produced instead of the 1064 keV γ, averaging 995 keV electron energy, provide an accurate calibration with considerable precision on the energy resolution. We usually combine that information with γ-ray Compton edges from ^{207}Bi, ^{137}Cs, and naturally occuring ^{40}K decay. This leaves a need to understand the energy calibration, resolution, and detector lineshape response at higher energies in our plastic scintillator.

Sources using (α,n) reactions on ^{9}Be and ^{13}C are neutron calibration standards, and both neutron spectra [2] and high-energy γ production [3] have been well-characterized in the literature. These sources are routinely used to calibrate large high-Z scintillators at γ energies 4.44 and 6.13 MeV produced by population of excited states of ^{12}C and ^{16}O, as the large pair production cross-sections and photopeak fractions can overcome events from inelastic scattering of fast neutrons and from thermal absorption, even in singles. We do not find in the literature use of these γ's to calibrate plastic scintillator. In our relatively large plastic scintillator, approximately 4% of 4.44 MeV γ's will Compton scatter, while approximately 0.5% will pair produce. We find here that timing and energy tags for both 511 keV γ's in back-to-back high-Z GAGG scintillators adequately selects the double-escape 3.42 MeV energy peak from the stopped pair of electron and positron. We address some of the challenges and backgrounds from neutron-produced events.

* behr@triumf.ca

II. EXPERIMENT

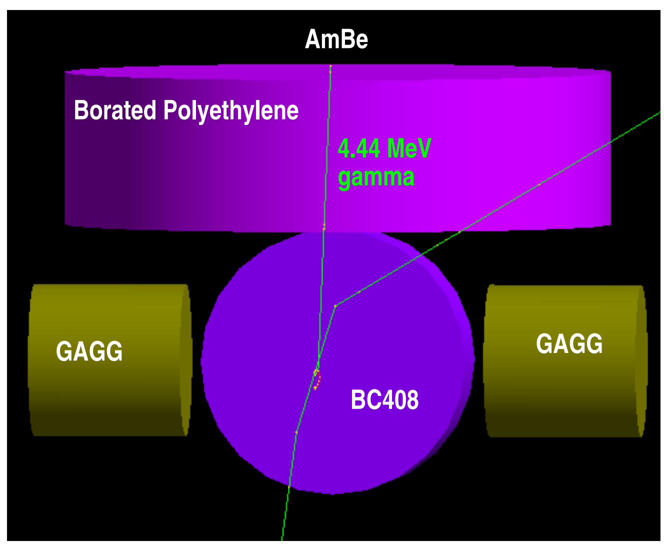

FIG. 1. GEANT4 model of the experimental geometry, showing 40x88 mm plastic scintillator, two 5x5cm GAGG scintillators, AmBe source location, and borated polyethylene shielding. A pair production event is shown where both annihilation γ's miss the GAGG's.

Figure 1 shows the geometry, with back-to-back 5x5 cm high-Z gadolinium aluminum garnet (GAGG) scintillators to detect the 511 keV γ's from pair production. The GAGG detectors have high photopeak fraction of

80-90% at 511 keV. The plastic scintillator is BC408. The sides are wrapped in about 10 layers of 0.003" thick teflon (i.e. about 0.75 mm). The front face is covered by nitrocellulose 0.12 mm thick, which in contrast with teflon [4, 5] is known to be highly reflective at this thickness [6], minimizing β energy loss and straggling. The photons are collected by a SensL ARRAYC-60035-64P-PCB 57x57mm silicon photomultiplier (SiPM), read out by an AiT Instruments ABL-ARRAY64PH board. The back surface area that is not covered by SiPM is covered in 0.75 mm thick teflon.

Large-area SiPM readout is used for all detectors. We caution that at first we saw large changes in bias currents of up to a factor of three in all three detectors in 12 hours of counting. Our ^{241}Am+Be (AmBe) source has 1x10^6 α decays/s and produces about 5 μSv/hr neutron doses at 0.3 m distance. Extrapolations from published SiPM effects from neutrons [7, 8] suggested that would not be an issue as we estimated 1/100 of fluences that produced damage, yet the bias currents stopped increasing when we added 5 cm of borated polyethelyne shielding. At least one prominent γ line in the GAGG's above 511 keV was reduced to negligible amounts by the neutron shielding. We were somewhat surprised that the very high neutron capture cross-sections of gadolinium isotopes are not producing overwhelming γ-ray backgrounds.

A. Coincidence data and cuts

1. *Relative timing*

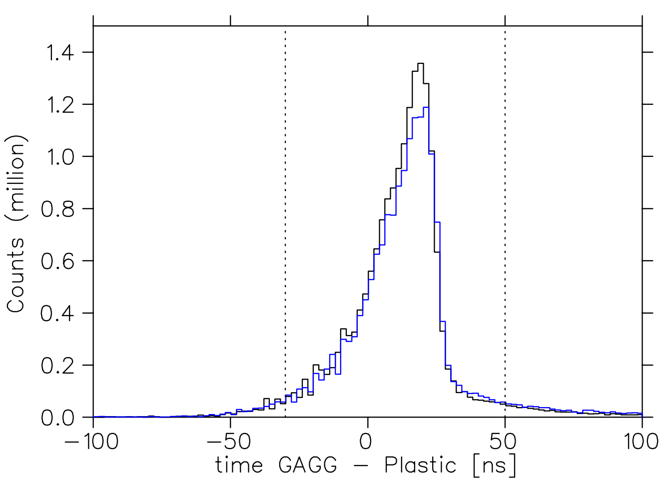

FIG. 2. Representative timing resolution of GAGG detectors with respect to the plastic scintillator (from 16 hr of data). Timing cut is indicated.

Conventional constant fraction timing produces relative timing spectra like the one in Figure 2, characterized by FWHM of 20 ns, inadequate to discriminate against possible background events from fast neutrons from the source. We find similar long tails at 10's of ns in natural background spectra without the source, so these tails are not necessarily coincidence events from the mulitple γ-rays expected from thermalized neutron capture. The large-area SiPM readout produces most of the 40$\pm$3 ns 10-90% risetime in the plastic, and part of the risetime of 86$\pm$3 ns in the GAGG, so in principle better coincidence timing could have produced plastic scintillator energy spectra with less background from source neutrons. Further spectra below are based on $\pm\ 2\sigma$ cuts in timing, taking about 95% of the total events. We have studied both more and less restrictive cuts, and find that tail and peak in the plastic are qualitatively similar.

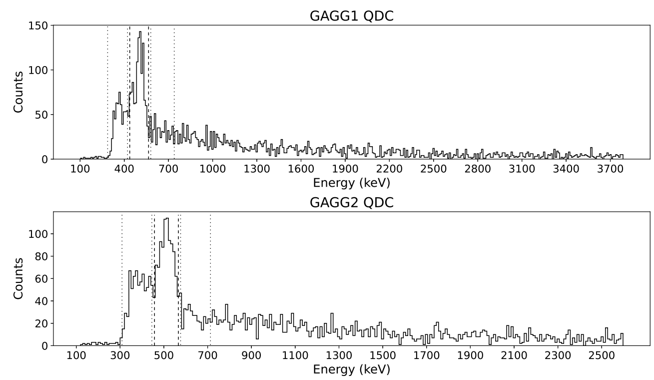

FIG. 3. Energy spectra of the GAGG detectors in timing coincidence with the plastic scintillator. Cuts of $\pm 2\sigma$ are shown around the annihilation peaks, along with background regions extending the next 5 σ above and below the signal.

2. γ *Energy*

The GAGG energy spectra in timing coincidence with the plastic are shown in Figure 3. Clear 511 keV annihilation photopeaks are seen.

There are more events at lower energy than the 511 keV photopeaks than would be expected from Compton edges. Thus by inspection there are also backgrounds continuous in energy extending under those photopeaks, both at lower and higher energy. We attribute these to possible events associated with neutrons not excluded by our coincidence timing cut. We will show the plastic scintillator energy spectrum for these background events in the next section.

B. Energy response of plastic scintillator at 3.42 MeV

The precision of the resulting high-energy calibration point at 3.42 MeV reveals imperfections in the detection system important for precision measurements.

Fig. 4 shows the measured plastic scintillator energy, in timing coincidence with GAGG and requiring 511 keV photopeaks in both GAGG detectors. The peak at 3.416

MeV is clear. This is the result of 88 hours of data collection. Note that singles spectra in the plastic (not shown) are overwhelmed by a continuum of neutron-produced events, with no photopeak seen at all.

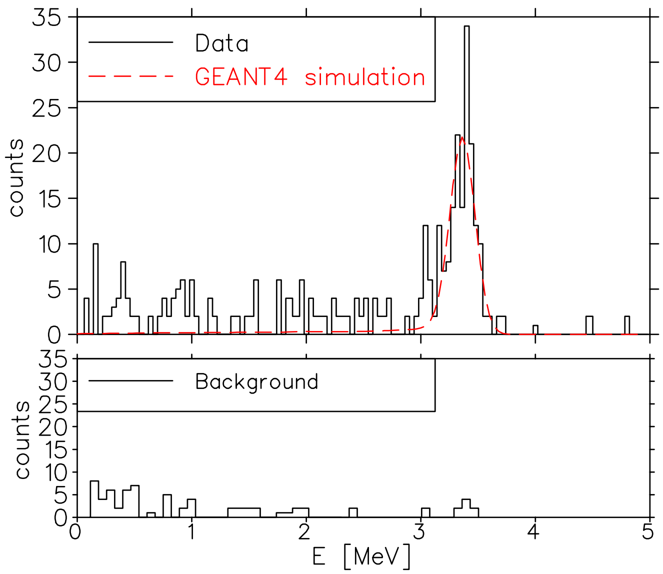

FIG. 4. Top: Energy spectrum of all plastic scintillator events in timing coincidence with both GAGG's, and requiring 511 keV energy in both GAGG's. Also shown is a GEANT4 simulation.
Bottom: The energy spectrum for background events in the plastic scintillator for events with 511 keV in one GAGG but more or less energy in the other GAGG.

1. *Energy calibration*

A fit to a linear energy calibration, Fig. 5, shows a systematic deviation. If all calibration data were used, the 3.42 MeV point would show 5% deviation from linearity with high statistical significance (25σ), while the fit has large χ^2. Such a nonlinearity seems unlikely given the known linearity of SiPM response at these relatively low count rates of a few 100 counts/sec.

A more physical explanation is to exclude the conversion electron from ^{207}Bi from the fit. The resulting fit of the γ sources is linear to better than 0.5% precision, and the residuals in Fig. 5 show a 3% deviation of the 995 keV electron. To show such a deviation, we require considerable precision and accuracy to extract the energy centroid produced by Compton edges, for which we use a differential method from the literature [9]. The fit still has a relatively high χ^2, with probability 0.5% that a random measurement would show a higher χ^2, which we attribute to difficulties of Compton edge extraction at precision $\sim$0.1%.

The higher gain for the conversion electron is consistent with 3% higher light collection from the first 5 mm of the plastic scintillator where the 995 keV electrons are stopped, compared to the γ-ray sources which uniformally illuminate the entire detector. This is roughly consistent with $\sim$5% nonuniformity of light collection we see in GEANT4 optical photon collection simulations for point sources at various radii in our geometry, given e.g. no light guide to help randomization. Note we have assumed 30 keV minimally ionizing energy loss in the 0.12 mm nitrocellulose front face reflector– increasing the average energy loss, or including a full simulation with energy straggling, would increase the discrepancy.

This systematic difference, revealed by the 3.42 MeV result, illustrates one challenge of using γ-ray based energy calibrations for β detection. Such nonuniformity of light collection seems quite small, and is understandable and acceptable to our experiments. Our experiments deliberately use the measured β spectrum as part of the energy calibration, to deduce the β energy dependence of spin-dependent asymmetries and angular correlations [1].

2. *Energy resolution achieved*

The FWHM resolution achieved at 3.42 MeV was 11.5±0.8% FWHM.

The energy resolution of the plastic scintillator system as a function of energy is shown in Fig. 5. We determine the energy resolution for Compton edges by convolution of a Gaussian with a GEANT4 simulation. We show a fit assuming photon statistics plus a constant offset. There is a substantial constant offset in energy resolution of 6%. Contributions to this could include random noise from dark current in the SiPM's, and of course the trivial possibility of noise contributions from our DAQ readout electronics.

Resolution for similar detector systems We have explored in a similar plastic scintillator setup, using this same C-type large-area SiPM, energy resolution at 995 keV as a function of overbias of the SiPM. Vendors' data indicates quantum efficiency increasing roughly linearly with overbias up to the 5 V overbias used for our data here, yet there is a possible tradeoff with the linear increase of dark current. We did not see much difference in energy resolution with overbias, and do not report this data here.

We note we have since improved resolution substantially at 995 keV from 14.3% to 10.2% for this C-type SiPM, and 9.3% for same-size J-type SiPM. Improvements came from an extra layer of dielectric mirror film outside the cylinder's teflon tape wrapping, better teflon reflection at front support and back of the plastic, and pressure applied to seal optical gel better between plastic and SiPM. The improved resolution is similar to what we have achieved previously with this scintillator size, a light guide to remove the sensor from the atom trap magnetic field, and a 12.5 cm diameter phototube with larger diameter than the light guide [10]. One would expect naively to get better energy resolution, as the quantum efficiency of SiPM's is substantially higher. The lower resolution may be partly due to imperfect light collection into the smaller 57x57 mm SiPM's, and partly due to SiPM dark current.

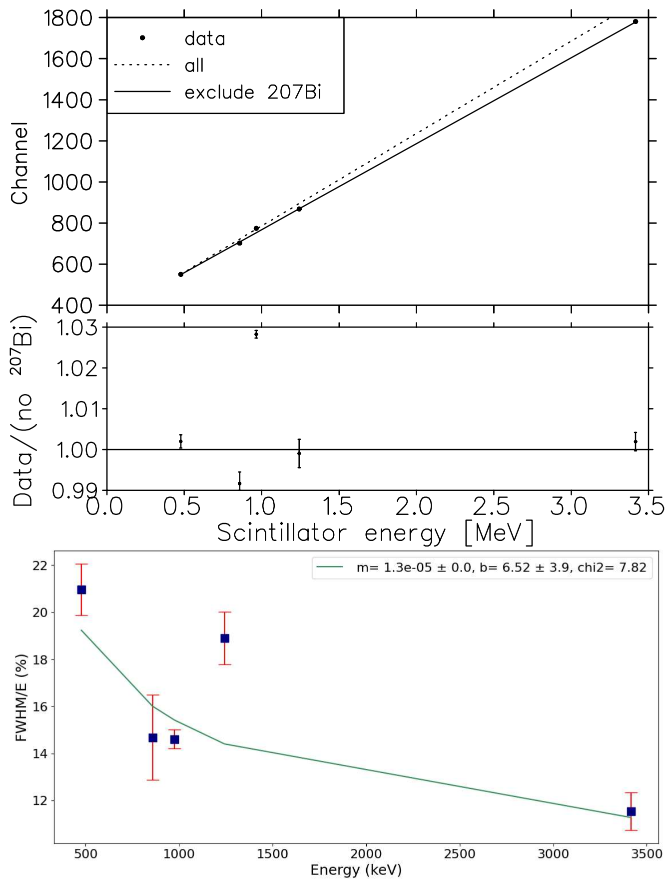

FIG. 5. Top: Deviations from linearity of the energy calibration. See text. Bottom: Energy resolution demonstrating photon statistics + constant component.

3. *Lineshape response and GEANT4 simulation*

A GEANT4 simulation for 4.44 MeV γ's is also shown in Fig. 4, using the emstandard$_{opt3}$ physics list. The simulation shows a tail on the response function, a combination of bremsstrahlung escape from the energetic e^+e^- pair and pairs created near the edge of the scintillator. The simulated tail below 90% of the peak has tail/peak ratio 0.13.

There are two qualitative disagreements between the simulation and the measured lineshape. There is a significant non-Gaussian distortion a few hundred keV below the peak which is not accounte for by the simulation. Possible sources of distortion include nonuniform light collection.

Also, the size of the long tail in energy is considerably greater in the data, especially at lowest scintillator energy. Also shown in Fig. 4 (Bottom) is the background energy spectrum from events with higher or lower energy in one GAGG scintillator, which show a substantially larger number of events at lower scintillator energy. So we attribute most of the low-energy tail to neutron-produced events mimicking the 511 coincidence without generating the 3.42 MeV peak. Although in principle modern simulations can include neutron-induced events, we do not consider the much greater complication here.

III. SUMMARY

We have measured 11.5±0.8% energy resolution of a 40x88 mm plastic scintillator at 3.42 MeV, using an AmBe neutron source that also produces 4.44 MeV γ's. The precision of the resulting calibration reveals challenges in such detection systems for precision measurements.

Comparison with lower-energy sources shows deviations from linearity of centroid energies. This is consistent with greater light collection by about 3% from the front center of the scintillator sampled by 1 MeV electrons.

Energy resolution improves with photon statistics, yet has a significant contribution constant with energy, possibly from SiPM dark current, electronic noise, and/or nouniform light collection.

The experimental lineshape tail provides an upper limit on its size. Quantitative understanding of the lineshape tail is limited by other backgrounds from the AmBe source, likely including events from fast neutrons that were not timed out.

Acknowledgements We acknowledge B.L. Lee for a previous generation of this experimental technique, and J.C. McNeil for a careful reading of the manuscript. Supported by RBC Foundation and the Natural Sciences and Engineering Research Council of Canada. TRIUMF receives federal funding via a contribution agreement through the National Research Council of Canada.

[1] "Precision Measurement of the β Asymmetry in Spin-Polarized ^{37}K Decay," B. Fenker, A. Gorelov, D. Melconian, J.A. Behr, M. Anholm, D. Ashery, R.S. Behling, I. Cohen, I. Craiciu, G. Gwinner, J. McNeil, M. Mehlman, K. Olchanski, P.D. Shidling, S. Smale, and C.L. Warner, Phys. Rev. Lett. **120**, 062502 (2018)

[2] "Neutron spectra of ^{241}Am/B, ^{241}Am/Be, ^{241}Am/F, ^{242}Cm/Be, ^{238}Pu/^{13}C, and ^{252}Cf isotopic neutron sources," E.A. Lorch, Int. J. Appl. Radiat. Isot. **24** 58 (1973)

[3] "A comparison of untagged gamma-ray and tagged-neutron yields from ^{241}AmBe and ^{238}PuBe sources," J.

Scherzinger, R. Al Jebali, J.R.M. Annand, K.G. Fissum, R.Hall-Wilton, S.Koufigar, N. Mauritzson, F. Messi, H. Perrey, and E. Rofors, Applied Radiation and Isotopes, **127** 98 (2017).

[4] "Dependence of polytetrafluoroethylene reflectance on thickness at visible and ultraviolet wavelengths in air," S. Ghosh, J. Haefner, J. Martín-Albo, R. Guenette, X. Li, A.A. Loya Villalpando, C. Burch, C. Adams, V. Álvarez, and L. Arazi, 2020 JINST 15 P11031

[5] "Reflectance dependence of polytetrafluoroethylene on thickness for xenon scintillation light," J.Haefner, A.Neff, M.Arthurs, E.Batista, D.Morton, M.Okunawo, K.Pushkin, A.Sander, S.Stephenson, Y.Wang, W.Lorenzon, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **856** 86 (2017)

[6] "Reflectivity Spectra for Commonly Used Reflectors," M. Janecek, IEEE Transactions on Nuclear Science, **59** 490 (2012)

[7] "Radiation damage in silicon photomultipliers exposed to neutron radiation," Yu. Musienko, A. Heering, R. Ruchti, M. Wayne, Yu. Andreev, A. Karneyeu, and V. Postoev, 2017 JINST 12 C07030

[8] "Neutron radiation damage and recovery studies of SiPMs," T. Tsang, T. Rao, S. Stoll and C. Woody, 2016 JINST 11 P12002

[9] "Differentiation method for localization of Compton edge in organic scintillation detectors," M.J. Safari, F. Abbasi Davani, H. Afarideh, arXiv:1610.09185.

[10] "Precise Measurement of the β-asymmetry in the Decay of Magneto-optically Trapped, Spin-Polarized ^{37}K," Benjamin Brown Fenker, Ph.D. thesis, Texas A&M, 2016.