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Low-cost pseudo-segmented neutron detectors using boron-loaded opaque scintillator

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Abstract.

Liquid scintillator detectors typically aim for high light collection by optimising their medium transparency to maximise photon collection by a single, large area, photo-sensor. Opaque scintillators, recently proposed as a radical alternative to this approach, instead focus on confining light as close as possible to its point of production by adopting scintillator mixtures with enhanced re-scattering probabilities. The use of fine arrays of optical fibres to read out scintillation light produced in this medium permits the imaging of particle interactions with extremely high resolution in a monolithic detector volume. We propose to expand on the opaque scintillator technique by producing an opaque detection medium from a neutron sensitive zinc-sulphide scintillator mixture doped with hexagonal boron nitride platelets. In this approach, we exploit the high light yield of zinc sulphide and the combined high neutron capture cross-section and reflective properties of boron nitride to demonstrate the potential for large volume neutron detection fabrication. This offers the potential for a low cost imaging neutron detector with position sensing and directional capabilities. In this paper, we discuss the progress achieved towards realising this novel neutron detector and potential for future development.

1 Introduction

Typically, liquid scintillators are optimised for high absorption and scattering lengths to preserve as much light as possible from neutron/gamma interactions and maximise detection efficiency, limiting primary scintillation fluors to those that can form stable transparent solutions. Recently, there have been several advances in the field of neutrino physics on the development of a new type of scintillating medium, known as opaque scintillator [1]. These opaque scintillators intentionally reduce the Rayleigh scattering length of the medium whilst maintaining a long absorption length. Modifying the scintillator in this way results in scintillation photons being repeatedly scattered with very low losses, confining the light close to its production point in a monolithic detector volume. By reading the scintillation light out with arrays of optical fibres it has been demonstrated that it is possible to produce a finely segmented detector in a monolithic scintillation medium with minimal detector deadspace.

In this work, we propose the development of new type of scintillator-based neutron detector utilising a novel Boron-Loaded Opaque Scintillator (BLOS) material. Over the past decade, research undertaken at the University of Sheffield [2] has investigated the use of unenriched hexagonal boron nitride to develop low-cost thermal neutron detectors as alternatives to traditional Helium-3 based systems. Many alternative thermal neutron sensors use ^6Li due to its high light output when combined with zinc sulphide scintillator (e.g. [3, 4]). However, producing neutron sensors using ^6Li is costly due to the need for

Isotope	^6Li	^{10}B
Natural abundance	7.50%	19.80%
n^{th} capture cross section	940 barns	3,842 barns
Capture products	$^4\text{He} + ^3\text{H} + 4.8 \text{ MeV}$	$^7\text{Li} + ^4\text{He} + 2.3 \text{ MeV} + 0.48 \text{ MeV}(\gamma) \text{ (94\%)} \\ ^7\text{Li} + ^4\text{He} + 2.8 \text{ MeV} \text{ (6\%)}$

Table 1: Comparison between ^6Li and ^{10}B as thermal neutron capture agents.

isotopic enrichment and its status as a controlled import/export material. In contrast, boron nitride is under no such restrictions and contains a higher natural abundance ($\sim 20\%$) of ^{10}B relative to the natural abundance of ^6Li . The thermal neutron capture cross section of ^{10}B is 3,842 barns, more than four times higher than that of ^6Li . The downside of using ^{10}B as the neutron capture agent is that the Q-value, and in turn the light output of the scintillator, is roughly halved relative to ^6Li for a neutron capture reaction.

A comparison between ^6Li and ^{10}B is shown in Table 1. Previous research has shown that thin layers of a mixture of boron nitride and zinc sulphide can be used to create thermal neutron sensitive scintillator with a reduced performance relative to traditional ^6LiF foils (for example EJ426 from Eljen) but at a fraction of the cost [2]. In this work we propose an alternative method to produce a large volume opaque scintillator based neutron detector. Instead of developing thin film scintillator technology we focus on the bulk loading of optically transparent resins with zinc sulphide, to add scintillation capability, and low particle size boron nitride platelets, to further reduce the scattering length and add neutron detection capabilities.

2 Fabrication of Boron-Loaded Opaque Scintillator

Hexagonal boron nitride is uniquely suited for opaque scintillator development, as boron nitride platelets are highly reflective ($\sim 95\%$ in the 400-500 nm region) and can easily be obtained in grain sizes as small as 200 nm (Pi-KEM). Both of these properties result in low-loss scattering in the emission wavelengths of zinc-sulphide when mixed together and added to a transparent suspension liquid. Boron nitride to zinc sulphide mixing ratios of between 1:3 and 1:4 by mass have been found to be optimal for producing neutron sensitive scintillator foils in [5]. This ensures that each ^{10}B atom is surrounded by zinc sulphide particles resulting in a high energy deposition in the scintillator by the neutron capture products. In this work we consider a similar BN:ZnS mixing ratio. Hexagonal boron nitride powder is mixed with silver activated zinc sulphide powder in a ratio of 1:4 to form the loading compound. Due to the inert nature of inorganic scintillators, a highly viscous liquid base is required to maintain the boron nitride zinc sulphide (BN:ZnS(Ag)) mixture in suspension. A bisphenol A-based clear epoxy casting resin was chosen for this purpose as it has high viscosity and high transparency across a broad range of wavelengths. To prepare prototype BLOS samples, 125 g of epoxy resin was measured out into a mixing pot. A quantity of BN:ZnS(Ag) powder for a desired loading concentration by mass was added to this epoxy casting resin before placing in a warm ultrasonic bath for 30 minutes followed by the solution being moved to a speed mixer running at 2,7000 RPM for two minutes. This was found to be sufficient to produce a well mixed BLOS solution that was relatively stable at room temperatures.

In total, four samples with loadings from 0.05% to 10% by mass relative to the amount of epoxy resin were produced. An example of the base resin mixture before and after 5% BLOS loading when exposed to UV light is shown in Figure 1. Unlike previous opaque scintillator studies, where impurities must be added to introduce the short scattering properties to an otherwise transparent medium, the BLOS mixture uses the natural reflectivity of the boron nitride to produce an opaque liquid. As expected the opacity of BLOS increases as the loading of BN:ZnS(Ag) increases, with transmission path length severely reducing as a small amount of BN:ZnS(Ag) is added as shown in Figure 1.

The optical transmission of the BLOS mixture was investigated using a Shimadzu 2700i UV-Vis spectrophotometer. Samples of both the undoped epoxy resin and BLOS were used to fill 1 cm cuvette cells for testing in the spectrophotometer. The spectrophotometer scans across a wavelength range from 200 nm to 800 nm and measures the percentage transmission through the sample. As can be seen in Figure 2, the epoxy resin shows good transparency across the wavelength range of the zinc sulphide

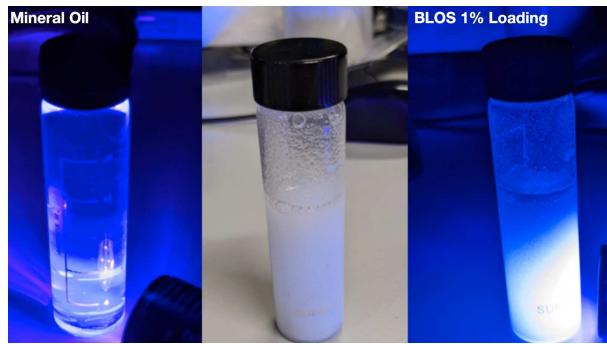


Figure 1: A vial containing an early BLOS formulation. Left: Base material under UV light. Middle: Base material with boron nitride zinc sulphide added. Right: BLOS under UV light showing light confinement within the vial.

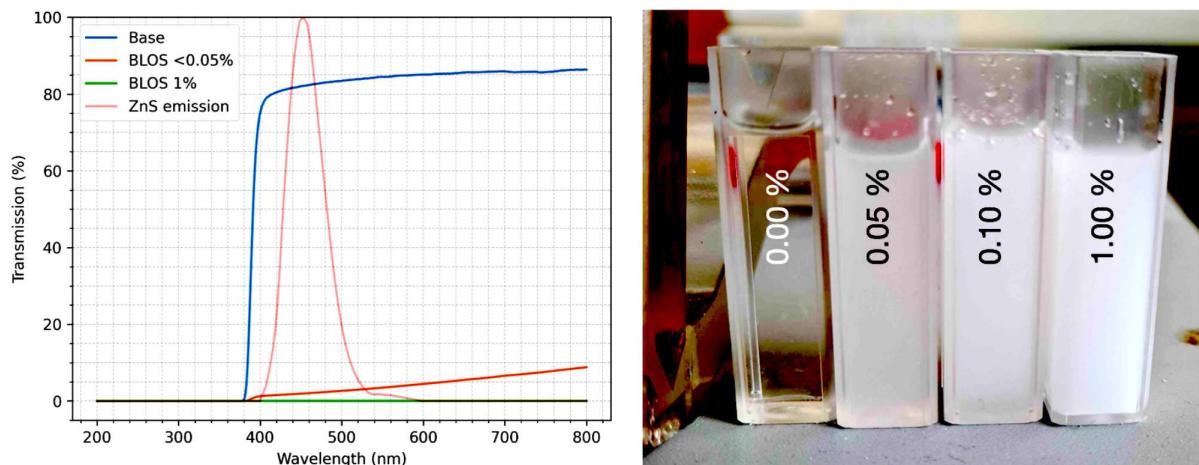


Figure 2: (Left) The light transmission measured through a 1 cm cuvette cell for the base undoped epoxy resin, 0.05% doped BLOS, and 1% doped BLOS. The zinc sulphide emission range is also shown. (Right) Cuvettes containing different BLOS concentrations. From left to right: 0.00% doping, 0.05% doping, 0.10% doping, 1.00% doping. The sample becomes more opaque with higher doping.

emission spectrum. It is important to minimise losses from the epoxy resin such that the vast majority of the optical effects are due to scattering on the boron nitride platelets rather than absorption in the base. Figure 2 also shows the effect on the light transmission through the 1 cm cell when the BN:ZnS(Ag) mixture is added. With the addition of less than 0.05% doping by mass, the transmission through the cell drops by an order of magnitude or more, depending on the wavelength, and with the addition of 1% doping, the transmission through the cell is negligible across the entire wavelength range. It is clear that at high doping the front face of the mixture in the cuvette is highly reflective and leads to a significant re-scattering of light. Further tests are ongoing to characterise the fraction of this loss in transmission that comes from re-scattering inside the material, and the fraction that is due to absorption of direct light when crossing the cuvette itself.

3 Prototype detector design

To demonstrate the use of BLOS for neutron detection a prototype system has been constructed. For this early detector prototype, a four-by-four array of 1.25 mm diameter BCF-91A wavelength-shifting single clad optical fibres [6] was used to extract light from a mixture of opaque scintillator. The fibres are inserted in a 1 cm horizontal grid pattern and are held in place using a custom-designed printed circuit boards (PCBs) utilising machined through holes to guide the fibres. A schematic of the fibre array

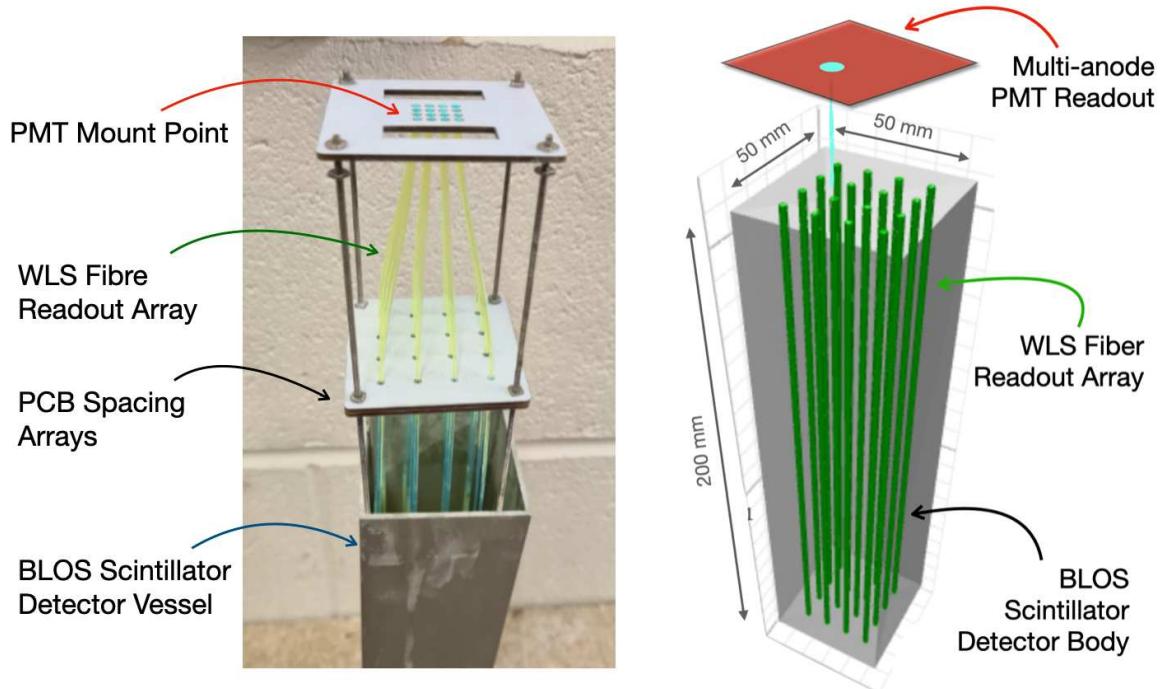


Figure 3: The prototype BLOS detector design showing optical fibres running through the length of the BLOS medium to a photosensor at the top.

design in shown in Figure 3. The use of PCBs as fibre guides is expected to support production of fibre assemblies in large quantities with good tolerance by PCB fabrication companies in the future. The fibres are used to guide collected light from the ZnS(Ag) scintillator to a Hamamatsu H11934 photomultiplier tube [7]. The PMT signals are digitised by a CAEN DT5725 [8] desktop waveform digitiser.

Pulse shape discrimination (PSD) can be used to separate gamma and neutron interactions in the BLOS detector. Neutron interactions result in a long, drawn-out emission of light from the zinc sulphide, whereas gamma interactions produce short, sharp peaks of light emission. We use a time-over-threshold algorithm to discriminate between neutrons and gammas, where neutrons will show a larger time-over-threshold value than gammas. The time window in which the algorithm is implemented spans from 50 ns prior to the first sample crossing threshold to 2 μ s after. The algorithm was chosen as it is straightforward to design and compute in programmable logic, and is lightweight enough to be replicated across many channels when multi-channel readout is implemented in the future. Figure 4 shows examples of waveforms resulting from gamma and neutron interactions in BLOS, and shows how the time-over-threshold can be used to discriminate between them.

We find that due to rescattering and absorption effects neutron interactions in BLOS often show a discrete train of pulses with reduced amplitude when compared to coupling wavelength shifting readout fibres to more traditional scintillator foils described in [2]. This is expected to be due to loss in collection efficiency on to the photosensors due to a closer matching of refractive indices between the fibre and surrounding liquid when capturing light in the wavelength shifting fibres, and transmissions losses at low wavelengths in the BLOS resin mixture. It is expected that pulse amplitude could be improved in the future using double clad wavelength shifting fibres, however for these early test the simple time-over-threshold PSD metric was found to still give a reasonable ability to discriminate neutron and gamma ray events. Figure 5 shows how neutrons and gammas produce two distinct populations in the time-over-threshold PSD plot, and that a cut on the time-over-threshold value can be used to discriminate between them.

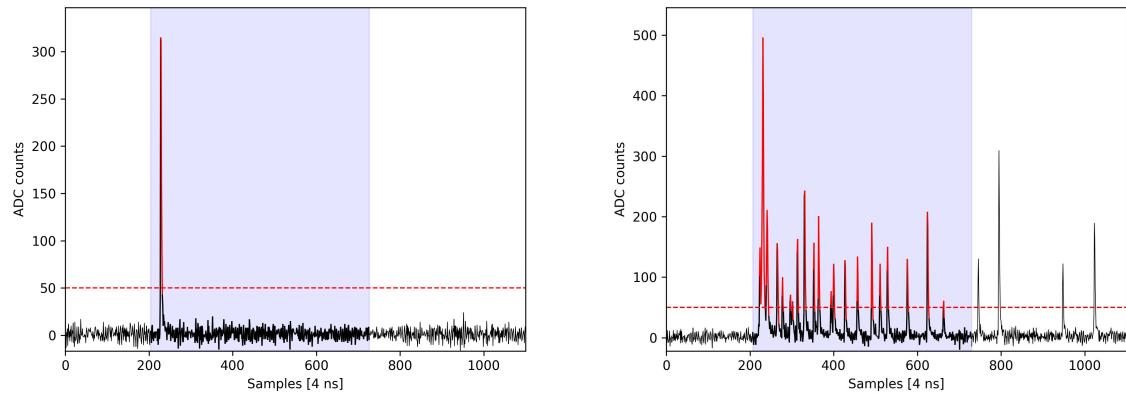


Figure 4: An example of a waveform resulting from a gamma interaction (left) and a neutron capture interaction (right) in BLOS. The regions of the waveform highlighted in red are above threshold (dashed red). Neutron pulses are identified by applying a cut on the time spent above threshold within a time window (shaded blue) after the initial threshold crossing.

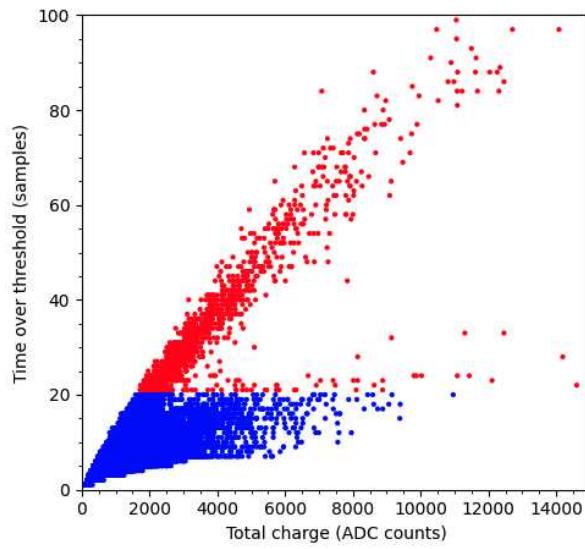


Figure 5: The time-over-threshold values for a BLOS sample irradiated with a moderated Cf-252 source. Events designated as neutrons, which passed a time-over-threshold cut of 20 samples, are shown in red while events that did not pass the cut are shown in blue.

4 Effect of BLOS loading on PSD

The BN:ZnS(Ag) dopant is directly responsible for both the neutron sensitivity and the opacity of the material. If the loading is too low, then the neutron capture efficiency will be reduced. However, if the loading is too high then it is expected that transmission losses between the capture vertex and the fibre resulting from the increased opacity will reduce the detection efficiency. Therefore, it is anticipated that there will be some optimal loading which balances the neutron capture efficiency with the optical properties of the medium for a given configuration of the optical fibre array.

To investigate the effects of the BN:ZnS(Ag) loading on the neutron detection capabilities of the BLOS detector, a series of BLOS samples with varying BN:ZnS(Ag) loadings were produced. The detector filled with a given BLOS sample was placed into a dark box and irradiated with a 1 MBq Cf-252 source moderated by high-density polyethylene. Data was acquired for one hour for each sample using a simple edge threshold trigger on the photomultiplier tube signal. The time-over-threshold analysis was applied to each waveform to produce the time-over-threshold PSD histograms for each sample that can be seen in Figure 6. These histograms show that there is an increase in the high time-over-threshold neutron population as the loading is increased from 0.05% to 1%. After this point, the neutron population begins to decline as the loading is increased further. More work is required to fully understand and quantify the absolute neutron detection efficiency as the BN:ZnS(Ag) loading varies and the optical effects that contribute towards the reduced neutron detection efficiency beyond 1% loading.

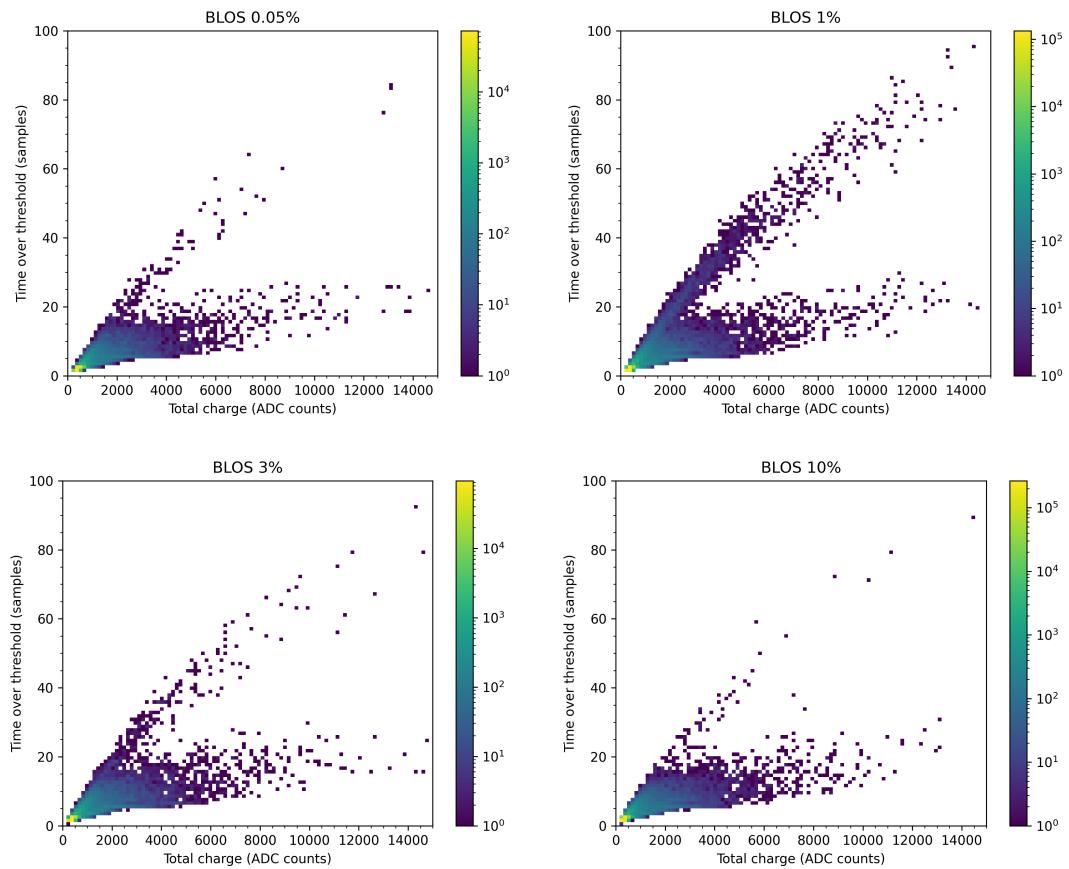


Figure 6: Time-over-threshold histograms demonstrating the effect of the BN:ZnS(Ag) loading on the neutron detection efficiency of the BLOS.

5 Future work

The BLOS concept is in the very early stages of development and more work is required to realise a positional and directional neutron sensor. We are actively investigating different base materials with

high viscosities and good transparencies in order to improve the stability of the medium and ensure that the boron nitride and zinc sulphide particles do not fall out of solution. One option is to cast the epoxy using hardener to produce a solid mass of detector volume. Alternatively, optical-grade liquid silicone rubbers such as Elastosil could be used which can be produced with a range of viscosities and optical properties.

In future work, a true multi-channel readout system will be developed. The monolithic photomultiplier tube included in the current setup will be replaced with a silicon photomultiplier array or a multi-anode photomultiplier tube, allowing each of the optical fibres to be read out by a separate photosensor. As the setup becomes more mature, the channel count will be increased, starting at 16 channels, then 32 channels, etc. In order to localise the position of the neutron capture within the BLOS volume, it will be important to measure the relative charge deposited from each fibre. Whilst this can be done in post-processing on a PC, we aim to implement real-time pulse processing on a Field-Programmable Gate Array (FPGA). The FPGA will use the same time-over-threshold algorithm as was demonstrated in section 4 to perform pulse shape discrimination on events in real time. Whilst other algorithms could potentially offer better discrimination capabilities, the time-over-threshold approach is straightforward to implement and can easily be scaled up to the much larger channel counts that are anticipated of future BLOS detectors.

Furthermore, as larger and larger BLOS prototypes are constructed, we expect to be able to investigate the self-moderating capabilities of the hydrogen-rich medium. This could provide sensitivity to higher energy neutrons and we would expect that neutron captures occurring closer to the middle of the volume will result from higher energy neutron thermalising within the medium. Simulation efforts are underway to investigate such effects.

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