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# Study and suppression of anomalous fast events in inorganic scintillators for dark matter searches

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## Abstract

The status of dark matter searches with inorganic scintillator detectors at Boulby mine is reviewed and the results of tests with a CsI(Tl) crystal are presented. The objectives of the latter experiment were to study anomalous fast events previously observed and to identify ways to remove this background. Clear indications were found that these events were due to surface contamination of

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crystals by alphas, probably from radon decay. A new array of unencapsulated NaI(Tl) crystals immersed either in liquid paraffin or pure nitrogen atmosphere is under construction at Boulby. Such an approach allows complete control of the surface of the crystals and the ability to remove any surface contamination. First data from the unencapsulated NaI(Tl) do not show the presence of anomalous fast events.

*Key words:* Scintillation detectors, Inorganic crystals, Dark matter, WIMP, Pulse shape analysis

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## 1. Introduction

The UK Dark Matter Collaboration (UKDMC) has been operating encapsulated NaI(Tl) detectors at the Boulby Mine underground site for several years [1]. Competitive limits on the flux of weakly interacting massive particles (WIMPs), that may constitute up to 90% of mass in the Galaxy, have been set by one of these detectors using pulse shape analysis (PSA) to distinguish between scintillation arising from background electron recoils and that due to nuclear recoils [2, 3]. Discrimination is possible because the sodium and iodine recoils expected from elastic scattering by WIMPs have faster mean pulse decay time than for electrons [4]. Traditionally, because NaI is hygroscopic, detectors are fabricated using an outer copper encapsulation with glued-in quartz windows plus additional thick (typically  $> 100$  mm) quartz lightguides to shield the crystal from photomultiplier activity. However, this design limits detector sensitivity because it prevents access to potential background sources on the crystal surfaces. The importance of NaI(Tl) surfaces has been highlighted recently by indications that they might be a source of anomalous fast time constant events seen so far at similar rates in many dark matter experiments with encapsulated NaI(Tl) crystals [1, 5, 6, 7]. Greater access would allow improved control of potential contaminants there and hence a possible reduction in such events, leading to greater sensitivity to WIMPs.

In this paper we report the results of a study of anomalous fast events with NaI(Tl) and CsI(Tl) detectors and a recipe to suppress their rate. We confirm also that unencapsulated NaI(Tl) crystals normally do not have this background.

## 2. Anomalous fast events

The observation of anomalous fast events in the UKDMC encapsulated NaI(Tl) detectors was first reported by Smith *et al.* [1]. These events are faster than typical electron recoil pulses and even faster than nuclear recoil pulses [1]. Figure 1a shows typical time constant distribution of events from one run with  $330 \text{ kg} \times \text{days}$  exposure of a 5.2 kg NaI(Tl) encapsulated crystal. The time constant distribution of events collected during calibration runs, when the crystal was irradiated by photons from a  $^{60}\text{Co}$  source, is shown in Figure 1b for comparison. Both distributions are fitted to a  $\log(\text{Gauss})$  function, shown as a solid line in Figure 1. The full data analysis procedure is described elsewhere [4, 6, 8, 9, 10]. A “bump” of anomalous fast events is clearly seen on the left edge of the distribution shown in Figure 1a. Note the absence of the bump in Figure 1b, which implies that the bump is not due to any non-uniformity of the crystal.

There is an excess of observed events over the  $\log(\text{Gauss})$  fit also at high values of time constant (see Figure 1). This can be explained assuming stochastic pile-up of single thermal photoelectrons [10], occasional afterpulses and fluorescence of the crystal after the scintillation pulse. The effect is seen for both “data” and “calibration” runs and does not interfere with the search for events faster than electron recoil events, such as nuclear recoils from WIMP interactions with matter or other kinds of fast events.

Figure 2 show time constant distributions for another run with the same encapsulated crystal (a) together with the results of a calibration run with a neutron source (b). Both neutron-induced and gamma-induced events are seen on the distribution plotted on Figure 2b. The fits to gamma-induced events (electron recoils) are shown by dashed curves. The fits to anomalous fast events (a) and neutron-induced events



(nuclear recoils) (b) are shown by dotted curves. From the comparison of Figures 2a and 2b we can conclude that anomalous fast events are faster than nuclear recoil events, expected from WIMP-nucleus interactions, and cannot be explained by WIMPs or neutron background.

Similar fast events with comparable rate have been seen also by the Saclay group [5]. One of the Saclay crystals [11], of size, growth technique, manufacturer, and housing similar to those used by the DAMA collaboration [12, 13], has been moved to Boulby (as a result of a collaboration between UKDMC and Saclay) and is currently collecting data. We confirm the presence of the population of fast events in this crystal with a rate similar to that seen in the UKDMC detectors (see also [11, 14] for discussion and Figure 6).

Smith *et al.* [1] suggested that the anomalous fast events could be due to MeV alphas. To account for the rate at low energies (10-100 keV) the alphas would need to deposit a small fraction of their initial energy at the crystal surface. Intrinsic bulk contamination of the crystal by uranium and thorium (measured to be at the level of about 0.1 ppb) is certainly not enough to explain the observed high rate at low energies. External incoming alphas from surrounding materials (PTFE – polytetrafluoroethylene, quartz windows) cannot easily explain the observed spectrum: fine tuning of model parameters, such as a dead layer of scintillator, is needed and a very high contamination of the material by uranium or thorium (about 1 ppm) is required as well. Moreover, the time constant of the incoming alphas is not matched well to that of the fast events [6].

Intrinsic surface contamination of the crystal by an alpha-emitting isotope has recently been discussed as a source of the anomalous fast events [7]. Recoiling nuclei from radon decay can be implanted into the crystal surface. This creates a thin (0.1-0.2 microns) alpha emitting layer. Although a high concentration of radioactive nuclei (0.1-1 ppm) is needed to account for the observed rate, the predicted spectrum agrees quite well with observations. Note that it is not known how such a large concentration of radioactive nuclei can appear on the surface of an encapsulated crystal.

Similar hypotheses on the source of the anomalous fast events have been suggested by Saclay groups [11, 15] and by Cooper *et al.* [16]. However, note that the hypothesis that anomalous events are due to  $^{214}\text{Po}$  decay [16] requires a constant supply of radon because of the short lives of isotopes decaying into  $^{214}\text{Po}$ .

If the source of fast events is indeed on the surface of the crystal, then it can be removed by polishing the surface. This is hard to do with encapsulated NaI(Tl) crystals but such an experiment can be done with CsI(Tl) providing it shows a similar rate of fast events. The advantages of CsI(Tl) crystals are: a) they are only slightly hygroscopic and can be easily handled; b) they show better discrimination capability between electron and nuclear recoils [8, 17]. (Note that CsI is generally ruled out for dark matter searches due to high intrinsic background).

### 3. Test with CsI(Tl) crystal

Tests were performed with an 0.8 kg CsI(Tl) Harshaw crystal previously studied in the laboratory to evaluate its characteristics such as quenching factor for recoils and discrimination power, relevant to dark matter searches. The results have been reported elsewhere [8].



The crystal was subsequently moved to the underground laboratory at Boulby and tested for background rate and anomalous fast events. In all tests we applied the standard procedure of pulse shape analysis adopted by the UK Dark Matter Collaboration for NaI(Tl) dark matter detectors [2, 6, 9, 8]. Pulses from both PMTs were integrated using a buffer circuit and then digitised using a LeCroy 9350A oscilloscope driven by a Macintosh computer running Labview-based data acquisition software. The digitised pulse shapes (10  $\mu$ s digitisation time) were passed to the computer and stored on disk. Final analysis was performed on the sum of the pulses from the two PMTs. Our standard procedure of data analysis involves the fitting of a single exponential to each integrated pulse to obtain the index of the exponent,  $\tau$ . Although scintillation pulses from CsI(Tl) have an additional second component [8], the pulses can nevertheless be well fitted by a single exponential if fits are restricted to data below 1500 ns. This fraction of the pulse contains the major contribution to the integrated pulse amplitude so that the distortion of the fit due to the presence of the second exponential at large time scales was found to be insignificant [8]. This approximation has the advantage that a three parameter fit can be used on each pulse and a simple discrimination parameter defined, rather than a considerably more complicated six parameter fit in the case of two decay constants. The free fit parameters used are: the time constant of the single exponent,  $\tau$ ; a normalisation constant and the start time of the pulse.

For each run the distribution of the number of events versus the time constant of the exponent ( $\tau$ ) was generated for a range of energy bins.  $\tau$ -distributions for each population of pulses can be approximated by a gaussian in  $\ln(\tau)$  (log(Gauss) function) [2, 6] (for a more detailed discussion of the distributions see [10] and references therein):

$$\frac{dN}{d\tau} = \frac{N_o}{\tau\sqrt{2\pi\ln w}} \times \exp \left[ \frac{-(\ln \tau - \ln \tau_o)^2}{2(\ln w)^2} \right] \quad (1)$$

The CsI(Tl)  $\tau$ -distributions were fitted with this gaussian in  $\ln(\tau)$  with the three free parameters: time constant  $\tau_o$ , width  $w$  and normalisation factor  $N_o$ . In experiments where a second population is seen (for example, nuclear recoils from a neutron source or anomalous fast events), the resulting  $\tau$ -distribution can be fitted with two log(Gauss) functions with the same width  $w$ .

In the low background conditions of the underground laboratory at Boulby, the CsI(Tl) crystal was found to show an anomalous population of fast events. Figure 3a shows time constant distribution of events with visible energy 30-50 keV together with a fit to a log(Gauss) function (14.3 kg  $\times$  days of exposure). The spectrum of these events presented in Figure 4 (crosses) has been traced up to MeV energies. The rate and shape of the spectrum are similar to those observed in the NaI(Tl) encapsulated detectors [1]. A peak at about 2.9 MeV corresponds to the 5.3 MeV alphas from  $^{210}\text{Po} \rightarrow ^{206}\text{Pb}$   $\alpha$ -decay, assuming a quenching factor for alphas of 0.55. (Scintillation efficiency of  $0.50 \pm 0.05$  for alphas was measured for this crystal with  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  sources). Note the absence of higher energy events which may be associated with the decay channels prior to  $^{210}\text{Po}$ . This is in contradiction to what was suggested in Ref. [16].

After 2 months of running at various dynamic ranges the crystal was removed, polished and put into a sealed vessel with nitrogen atmosphere. After polishing the crystal was exposed to air for only a few hours during the installation procedure.

The subsequent runs revealed a decrease in the rate of fast events by about a factor



of 4 (squares in Figure 4). The first two points below 100 keV show an upper limit to the rate. An accurate measurement of the rate at these energies is difficult because of the small mass of the crystal and the high rate of  $\gamma$ -background observed due to internal contamination of  $^{137}\text{Cs}$ . The time constant distribution for events of 30-50 keV after polishing is shown in Figure 3b (22.3 kg  $\times$  days of exposure). It can be seen that the rate of anomalous fast events is significantly reduced (see Figure 3a), though not completely suppressed probably due to the difficulty of removing the hard surface layer of CsI.

17 high-energy events (visible energy of 5-6 MeV) were also detected during the first day after polishing. These are double-pulse events where the first pulse corresponds to the  $\beta$ -decay of  $^{212}\text{Bi}$  (half-life 1 hour) and the second due to the  $\alpha$ -decay of  $^{212}\text{Po}$  (half-life 0.3  $\mu\text{s}$ ). These events are probably caused by contamination of the crystal surface during installation. No more of these events were seen after the first day (half-life of parent isotope  $^{212}\text{Pb}$  is 10.6 hours). Visible energy of these double-pulse events agrees with the assumption that the scintillation efficiency of alphas is about 0.5.

Only one prominent peak is seen in the spectra shown in Figure 4. The peak is probably due to  $^{210}\text{Po} \rightarrow ^{206}\text{Pb}$   $\alpha$ -decay (5.3 MeV  $\alpha$ s). No decay chains  $^{222}\text{Rn} \rightarrow ^{218}\text{Po} \rightarrow ^{214}\text{Pb}$  or  $^{224}\text{Ra} \rightarrow ^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb}$  were seen before or after polishing. This suggests that the concentration of U, Th and Ra in the bulk of the crystal is very low (less than 0.1 ppb).

At least several months of exposure to Rn is needed to explain the rate of  $\alpha$ -events in CsI(Tl) and NaI(Tl) detectors. This is not surprising for an unencapsulated CsI crystal but is hard to explain for NaI sealed detectors.

#### 4. Array of unencapsulated NaI(Tl) detectors - NAIAD

The results obtained with the CsI(Tl) crystal at Boulby clearly indicate the importance of having access to the crystal surface for polishing and control. Such access can be granted by running unencapsulated crystals in high purity mineral oil or dry gas inside sealed plastic or copper vessels. Laboratory tests have shown also that high light yield (up to 10 photoelectrons per keV) can be reached with the aforementioned detectors [9, 18].

Since 1999 the UKDMC has been developing a programme to run several unencapsulated NaI(Tl) crystals mounted in an array – NAIAD (NAI Advanced Detector). The NAIAD array is designed to be flexible enough to allow various modes of operation with crystals. To date two types of module have been constructed: a “vertical” module filled with high purity mineral oil to protect crystals from moisture, and a “horizontal” module in which either an oil or dry nitrogen is used around the crystal. Operation of an encapsulated crystal is also possible in the horizontal module. Design details and predictions of sensitivity to WIMPs are given in Ref. [9].

The first vertical module of NAIAD has been running since February 2000. It contains a 14 cm diameter  $\times$  15 cm length crystal (termed DM74) with mass of about 8.5 kg. The crystal was polished before installation. The total exposure (excluding calibration runs) is 996 kg  $\times$  days. Figure 5 shows a typical distribution of time constants for events with 35-40 keV visible energy together with a fit to a log(Gauss) function. As with the polished CsI(Tl) crystal the rate of anomalous fast events is greatly suppressed. The calculated limit on the rate of anomalous fast events as a



function of visible energy is shown in Figure 6 together with the energy spectra of fast events measured in typical encapsulated crystals.

A second (horizontal) module containing 4 kg unencapsulated and polished crystal in nitrogen (DM72) also does not show the presence of fast events. This second detector is currently running underground at Boulby. Analysis of data from both crystals in terms of limits on the WIMP-nucleon and WIMP-proton cross-sections is in progress.

## 5. Pulse shape analysis versus annual modulation

Pulse shape analysis (PSA) is not the only technique used with NaI(Tl) detectors for dark matter searches. The DAMA group [12] searches for an annual modulation in the background counting rate of their NaI(Tl) array without PSA (PSA is used only to discriminate between scintillation pulses and PMT noise). Evidence for such an annual modulation in the background rate has been reported by DAMA (see [12] and references therein) indicating a possible signal from WIMPs.

It would appear that the DAMA group could, in fact, confirm or exclude this possibility using pulse shape analysis. In Ref. [12] DAMA presented results of the annual modulation analysis (positive signal) together with previous limits on WIMP-nucleon cross-section obtained with PSA. All sets of data (DAMA/NaI-0 analysed using PSA and DAMA/NaI-1 – DAMA/NaI-4 analysed using annual modulation) were obtained with the same experimental set-up and under similar conditions such as background rate etc. Pulse shape analysis applied to the first data set (DAMA/NaI-0) allowed DAMA to put limits on WIMP-nucleon interactions of the order of  $(5 - 6) \times 10^{-6}$  pb for 50-100 GeV mass WIMPs with halo density of  $0.3 \text{ GeV/cm}^3$  (see [12, 13] for details and halo parameters used). This limit was obtained with  $4123.2 \text{ kg} \times \text{days}$  exposure. The subsequent data sets (DAMA/NaI-1 – DAMA/NaI-4) totaling  $57986 \text{ kg} \times \text{days}$  showed a positive signal at  $4 \sigma$  confidence level using annual modulation analysis without PSA. WIMP parameters derived from this analysis are:  $M_W = (52^{+10}_{-8}) \text{ GeV}$  and cross-section  $\sigma_p = (7.2^{+0.4}_{-0.9}) \times 10^{-6} \text{ pb}$  with the same halo parameters [12]. Simple statistical considerations show that PSA applied to this 15 times larger exposure from all five data sets (DAMA/NaI-0 – DAMA/NaI-4) compared to the first one (DAMA/NaI-0) could yield a limit on the WIMP-nucleon cross-section improved by a factor of 3.9. Such an analysis would allow DAMA either to confirm the modulated signal or to exclude practically the whole region of parameters that they derive from the observed modulated signal (see figures 3 and 4 in Ref. [12] for the DAMA allowed region of WIMP parameters). The same statistical considerations suggest that the signal reported in Ref. [19] for the period DAMA/NaI-1 with  $4549 \text{ kg} \times \text{days}$  should have been at a  $1.1 \sigma$  confidence level, to be consistent with the signal observed at  $4 \sigma$  confidence level with  $57986 \text{ kg} \times \text{days}$  exposure.

## 6. Conclusions

Tests with a CsI(Tl) and unencapsulated NaI(Tl) crystals have shown that anomalous fast events seen in several NaI(Tl) detectors at Boulby were probably due to surface  $\alpha$ s. Radioactive  $\alpha$ -emitting isotopes had likely been implanted into crystal surfaces by radon decay. Polishing the crystal surfaces removed a major part of the fast events. A new array of unencapsulated NaI(Tl) crystals (NAIAD) is being installed in the underground laboratory at Boulby. Data from the first of these modules do not reveal



anomalous fast events.

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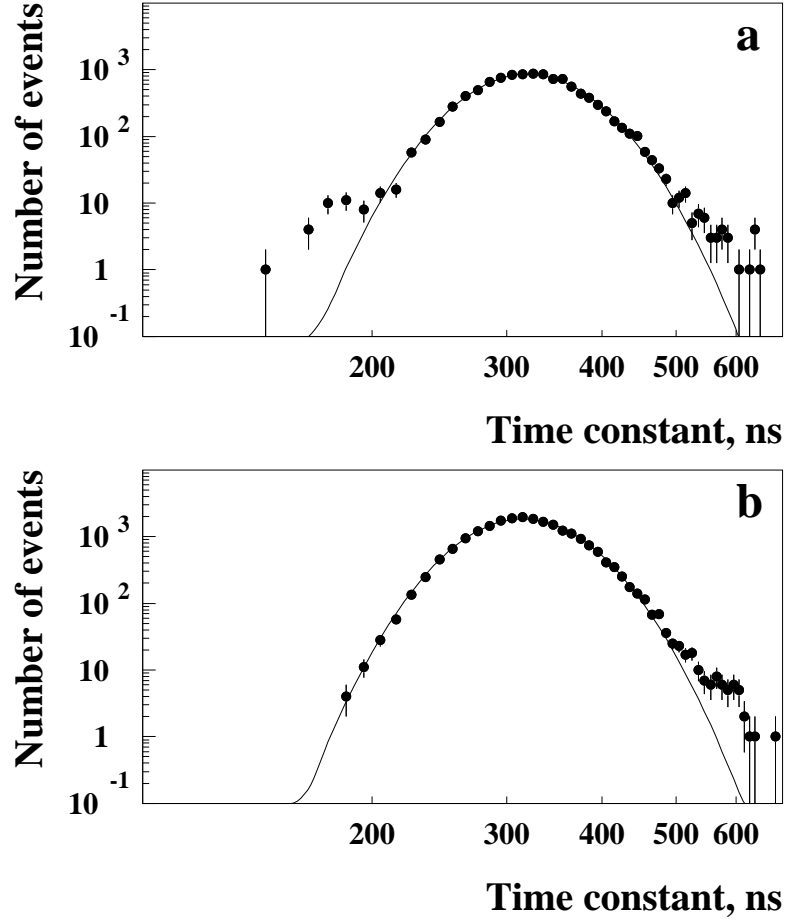


Figure 1: a) Time constant distribution for events with visible energy 35-40 keV from one encapsulated NaI(Tl) detector; b) similar distribution for Compton events from a gamma source. Solid curves show fits to Gaussian distributions on a logarithmic scale ( $\log(\text{Gauss})$ -function).



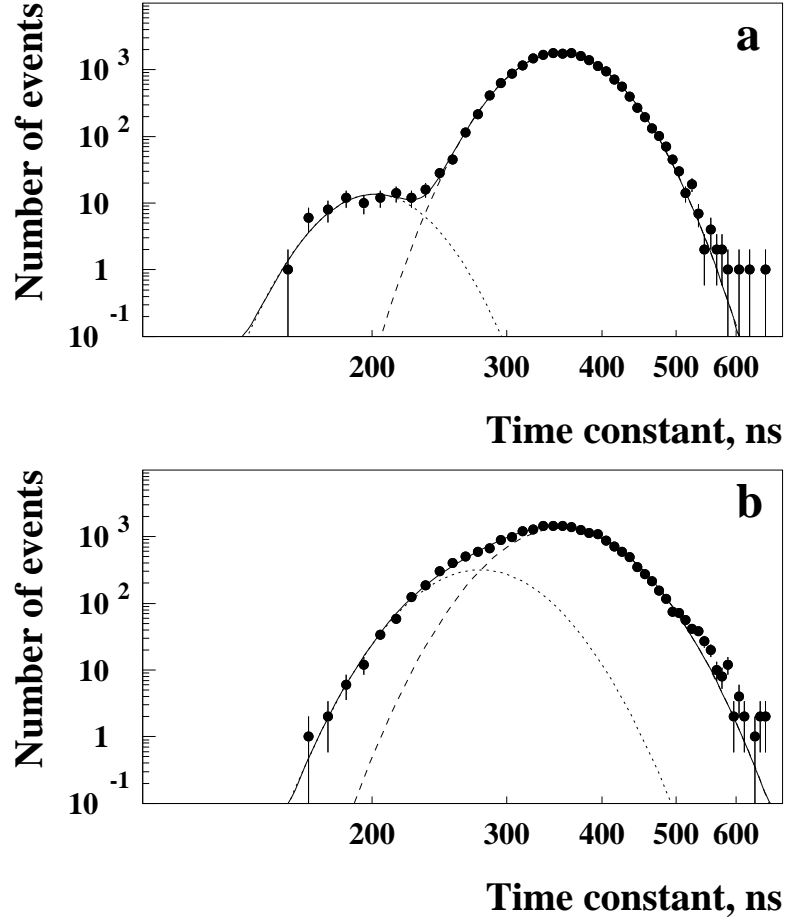


Figure 2: a) Time constant distribution for events with visible energy 40-45 keV from encapsulated NaI(Tl) detector; b) similar distribution for calibration run with neutron source. Solid curves show fits to a sum of 2 log(Gauss)-functions. Dashed curves show fits to gamma-induced events. Fits to anomalous fast events (a) and neutron-induced events (b) are plotted by dotted curves.



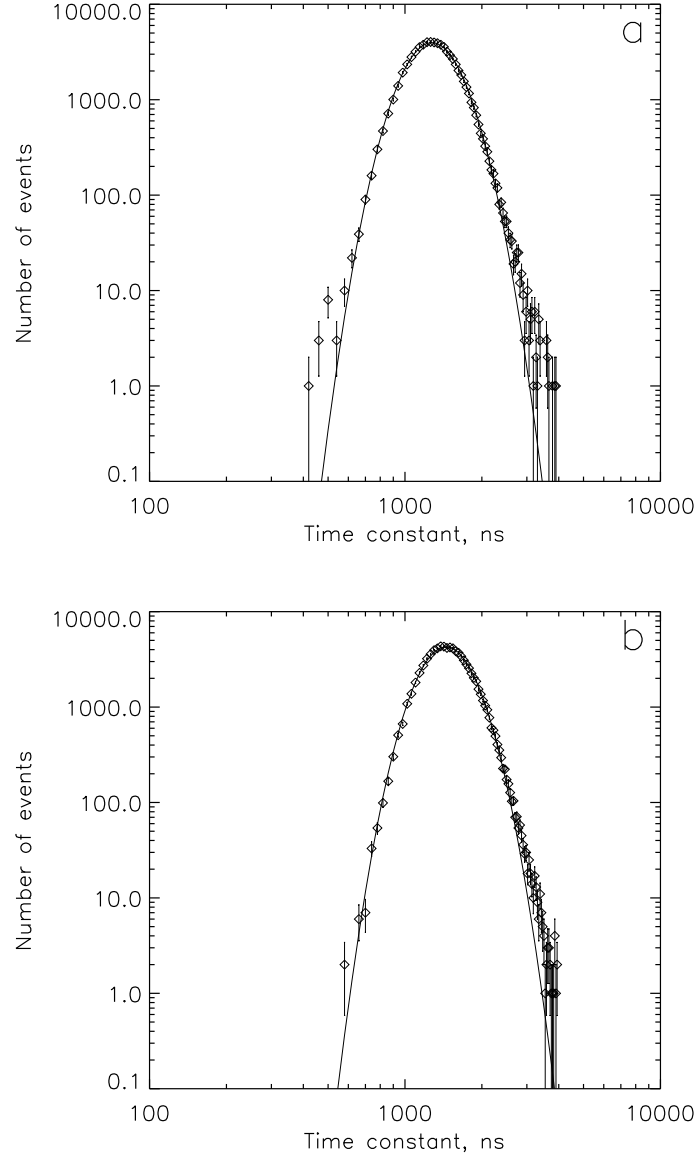


Figure 3: a) Time constant distribution for events with visible energy 30-50 keV from the CsI(Tl) crystal; b) similar distribution after crystal polishing. Solid curves show fits to a log(Gauss)-function.



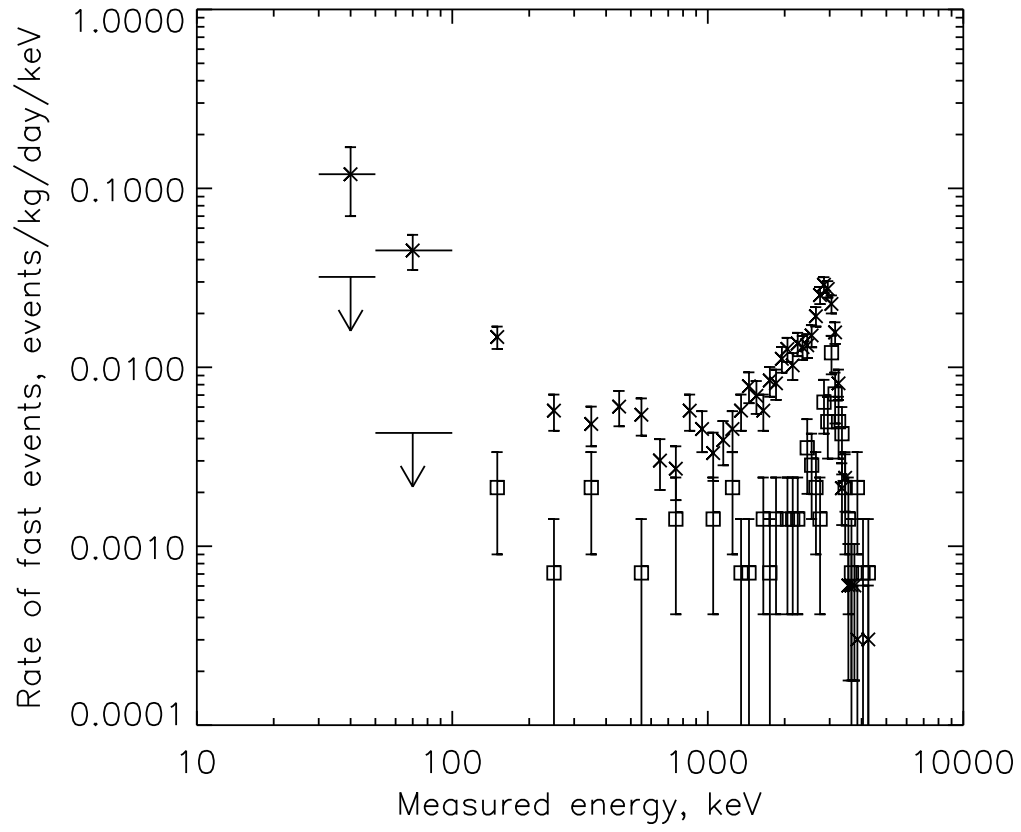


Figure 4: Rate of fast events ( $\alpha$ s) in CsI(Tl) crystal before (crosses) and after (squares) polishing. The first two points after polishing show limits at 90% confidence level.



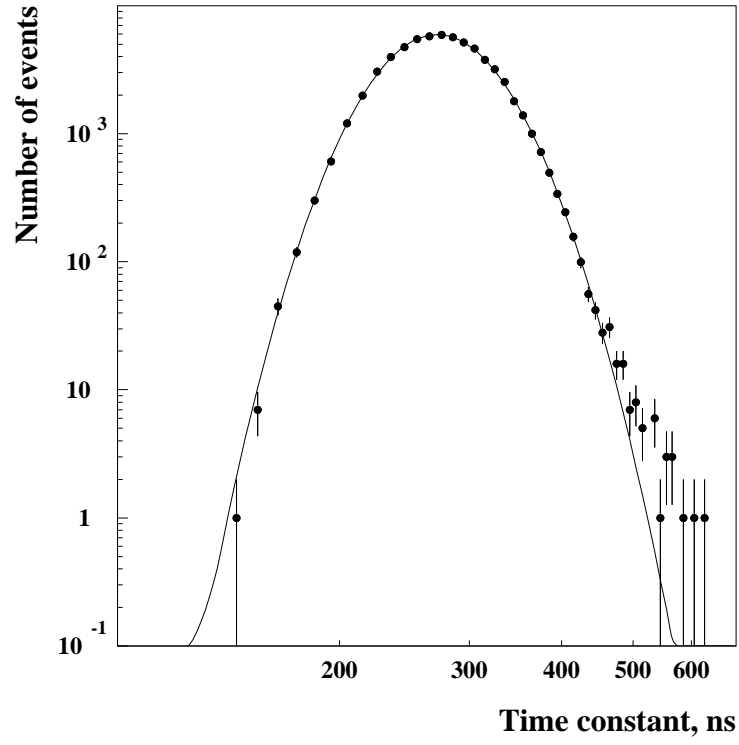


Figure 5: Time constant distribution for events with visible energy 35-40 keV from the first NAIAD module (DM74). Solid curve shows a fit to a log(Gauss)-function.



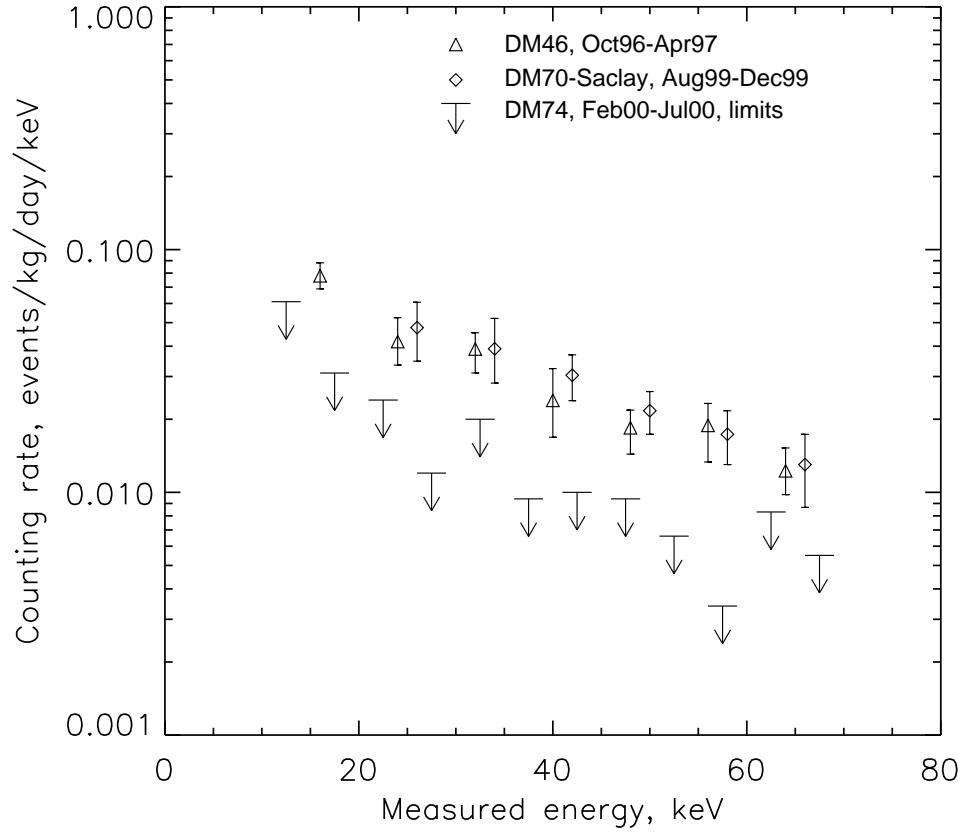


Figure 6: Rate of anomalous fast events in NaI(Tl) encapsulated crystals and limits on the rate of these events in the unencapsulated crystal (DM74): triangles – UKDMC DM46 crystal; diamonds – Saclay crystal, also named DM70 (see also [11, 14]); arrows – limits at 90% confidence level from DM74 data.