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A possible mechanism for anomalous pulses observed in sodium iodide crystals

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Abstract

Low energy pulses with anomalously low time constant have been observed recently in sodium iodide crystals. A model is presented based on partial energy deposits from alpha particles passing out of the crystal from a thin contaminated layer, which would generate events of similar characteristic. The predicted $1/E^2$ form of the spectrum is consistent with observation. The required level of contamination is estimated both for U/Th impurities and for radon decay products but is found to be higher than would be expected from the crystal fabrication process and history. Possible experimental tests of the model are suggested. © 2000 Published by Elsevier Science B.V.

1. Introduction

NaI scintillating crystals have been used extensively for particle detection and discrimination for over 40 years [1] but only in recent years have attempts been made to extend this technique to much higher sensitivity at low energies, to search for rare neutral particle interactions from Galactic dark matter, with further proposals for solar neutrino, and neutrino magnetic moment experiments [2].

In searching for low energy nuclear recoils by low background underground experiments in the Boulby Mine, a new population of unidentified events was observed in sodium iodide crystals [3,4]. These showed pulse time constants about $62 \pm 2\%$ of the time constants of gamma pulses of the same energy (Fig. 1). This is significantly shorter than that of the nuclear recoil pulses produced by neutron scattering, which are at $73 \pm 2\%$ of the gamma time constant. Because of the finite width of the time constant distributions these events may overlap into the low energy nuclear recoil region and form an additional background for dark matter experiments. The existence of these unidentified events was subsequently independently confirmed in experiments at the Modane laboratory [5].

It was conjectured in [3] that these events could in principle be alpha interactions, which at MeV energies are seen to give pulse time constants $\sim$ 0.7 of the gamma time constant, with a trend consistent with the possibility of a lower ratio at keV energies. However, the known alpha activity in the crystal is insufficient to account for the observed event rates. Fig. 2 shows the differential energy spectrum of both the unidentified events and the MeV-range alphas from U/Th contamination. The total rate of the low energy events < 100 keV has a value $\sim$ 6% of the total rate of alphas > 1 MeV from intrinsic U/Th

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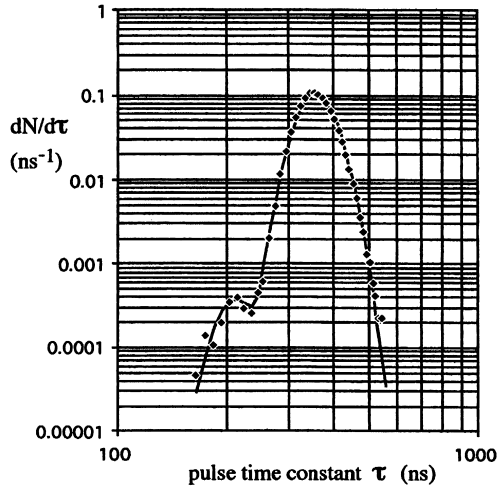

Fig. 1. Distribution of pulse time constants for a 5 kg low background NaI crystal (data for energy range 50–70 keV) showing main distribution of gamma pulses centred on 350 ns and additional unidentified population of pulses at 210 ns (from Ref. [3]).

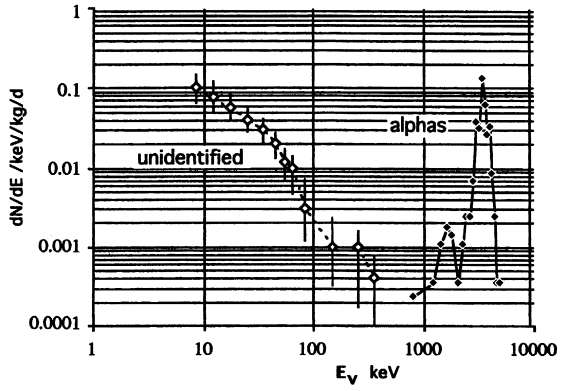

Fig. 2. Rate versus energy of unidentified low energy pulses in NaI crystal, together with measured spectrum of intrinsic alpha events above 1 MeV, resulting from bulk U/Th contamination [3].

contamination. This is too high to result simply from an 'escape' loss from the latter, since a surface thickness comparable to the ~ 30 μm alpha projected range represents only $\sim 10^{-3}$ of the crystal volume. However, a high surface contamination might still be responsible. Laboratory tests in the UK [6] have confirmed that the time constant of the anomalous pulses does not match that of beta particles (which is similar to that of gammas), but that alpha particles depositing energy near the surface can give pulse time constants shorter than that of nuclear recoils [7].[1]

Thus, on the hypothesis that alpha interactions might be responsible, an explanation of the unidentified events must also account for (a) the low energy deposition of these events. (b) the approximately $1/E^2$ behaviour of the energy spectrum. (c) the relatively high rate of these events (several per day in a 5 kg crystal)

In this letter we put forward a model which can account for both properties (a) and (b) through a finite-thickness layer of alpha-emitting impurity near the crystal surface, and to estimate the level of contamination required to account for (c) either in terms of Radon decay products or from a high surface contamination of U or Th.

2. Model and energy spectrum

Consider a thin layer ($\ll$ alpha range) of alpha-emitting contamination extending from the crystal surface into the crystal. Alphas emitted towards the interior of the crystal will deposit their full energy, while alphas emitted towards the surface will leave

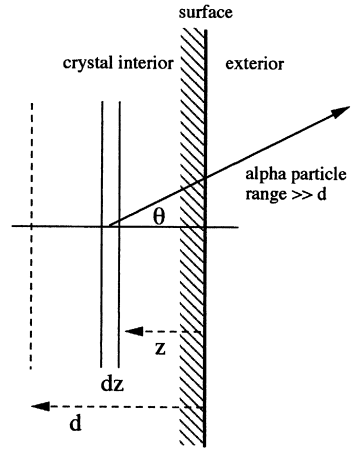

Fig. 3. Notation for estimating energy loss distribution for alpha particles passing out of crystal from surface layer $\ll$ alpha range.

[1] Tests at Sheffield Univeristy subsequent to [7] have shown surface alpha interactions with time constants down to 0.6 of the gamma time constant (N.J.C. Spooner, personal communication).

the surface depositing part of their energy. The energy spectrum can be derived as follows:

Referring to Fig. 3, alphas emitted with energy E_α at a distance z from the surface and angle θ will deposit on average an observed energy

$$E \approx E_\alpha(z/R_\alpha)(1/\cos\theta), \tag{1}$$

where R_α is the full projected range of the alpha in the material. The linear relation (1) is expected to be a good approximation below 100 keV, which is less than 2% of the alpha energy. Thus, for $\Delta n_\alpha(z)$ alphas emitted in a small volume element at z, integrating over azimuth, and putting $q = \cos\theta$, we have

$$\mathrm{d}n = 0.5\Delta n_\alpha(z)\,\mathrm{d}q, \tag{2}$$

$$\mathrm{d}E = E_\alpha(z/R_\alpha)(1/q^2)\,\mathrm{d}q, \quad q < 1, \tag{3}$$

giving the differential spectrum at distance z

$$\mathrm{d}n(z)/\mathrm{d}E = \Delta n_\alpha(z)\,(E_\alpha/R_\alpha)(z/2E^2), \tag{4}$$

with the constraint

$$E \geqslant E_\alpha(z/R_\alpha) \quad \text{or } z \leqslant R_\alpha(E/E_\alpha). \tag{5}$$

For a slab of thickness $\mathrm{d}z$ (cm), area S (cm^2), density ρ (g cm^{-3}) containing alpha activity $B_\alpha(z)$ (Bq g^{-1})

$$\Delta n_\alpha(z) = B_\alpha(z)\,\rho S\,\mathrm{d}z. \tag{6}$$

The differential spectrum for a finite thickness is then

$$\mathrm{d}n/\mathrm{d}E = (\rho S/2)(E_\alpha/R_\alpha)(1/E^2)\int B_\alpha(z)\,z\,\mathrm{d}z,$$

$$0 \leqslant z \leqslant R_\alpha(E/E_\alpha). \tag{7}$$

Consider the case of $B_\alpha(z) = \text{constant} = B_\alpha$ from $z = 0$ to $z = d$. The spectrum has two regions, corresponding to the restriction $z \leqslant R_\alpha(E/E_\alpha)$:

(i) If $E < E_\alpha d/R_\alpha$ the integral extends from $z = 0$ to $z = R_\alpha(E/E_\alpha)$, for which (7) becomes

$$\mathrm{d}n/\mathrm{d}E = (B_\alpha \rho S/4)(R_\alpha/E_\alpha) = \text{constant}. \tag{8}$$

(ii) If $E > E_\alpha d/R_\alpha$ the upper limit for z becomes d (the assumed thickness of the alpha emitting region). In this case (7) becomes

$$\mathrm{d}n/\mathrm{d}E = (B_\alpha \rho S/4)(R_\alpha/E_\alpha)(d/R_\alpha)^2 \times (E_\alpha/E)^2, \tag{9}$$

which joins to (8) at the point

$$E = E_\alpha d/R_\alpha. \tag{10}$$

For a scintillator calibrated in the usual way with gammas (electron recoils) the apparent deposited energy for alphas will be in general smaller, with the observed 'visible energy' E_v related to the true energy E by $E_v = f_\alpha E$ where f_α is the relative scintillation efficiency. We assume the observed high energy value $f_\alpha \approx 0.7$ for alphas in NaI (with the emitted alphas all in the MeV range, any energy dependence of f_α will have only a marginal effect). Then, for the observed differential spectra, (8) and (9) are modified to

$$\mathrm{d}n/\mathrm{d}E_v = (B_\alpha \rho S/4)(R_\alpha/E_\alpha)(1/f_\alpha) = \text{constant}. \tag{11}$$

$$\mathrm{d}n/\mathrm{d}E_v = (B_\alpha \rho S/4)(R_\alpha/E_\alpha)(f_\alpha)(d/R_\alpha)^2 \times (E_\alpha/E_v)^2. \tag{12}$$

Thus an alpha-emitting layer of constant B_α and thickness $d < R_\alpha$ would give a spectrum of low energy events from escaping alphas which has a flat region followed by a region falling as $1/E_v^2$. An example of the behaviour of (8), (9) and (10) is shown by curve (a) in Fig. 4. The junction of the two regions would in practice be smoothed out by range straggling. A further small correction would result from inclusion of the nuclear recoil energy. Although this is of order 100 keV, its observed value will be substantially reduced by the quenching factor f_A for the heavy nucleus. The value of f_A in general falls with increasing A [8,9] having the known values 0.7, 0.3, 0.08 for $A = 4$, 23, 127, extrapolating to < 0.05 for $A \sim 230$. Thus the recoil correction should be < 5 keV, modifying the spectrum by less than 10–20% and again smoothed by straggling effects.

When B_α varies with z, the integral (7) is modified. The constant and $1/E^2$ regions remain, but the transition is continuous rather than at a single point, as indicated by curve (b) (which refers to the specific example calculated below for Radon decay products). Of course, further modifications would result from

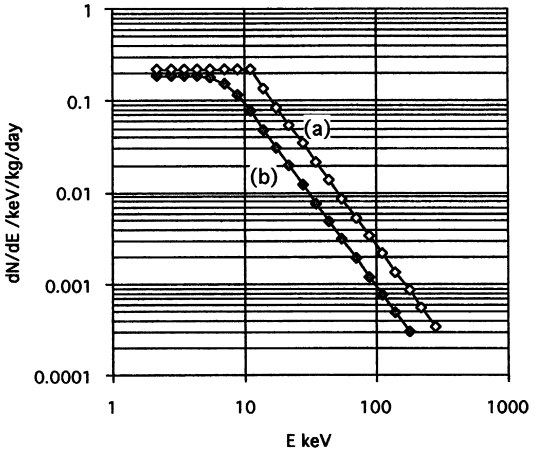

Fig. 4. Examples of differential energy spectrum for alpha particles leaving crystal from thin surface layer, showing transition to $1/E^2$ behaviour with increasing E. Curve (a) typical spectrum from layer with uniform concentration Curve (b) spectrum from recoil concentration distribution of Fig. 5.

variations in B_α over the surface S, but the overall $(1/E)^2$ dependence would remain.

3. Required concentration of U or Th

In comparing the predictions of Fig. 4 with the observed spectrum of Fig. 2, the horizontal position of the spectrum can be matched by varying the layer thickness d, while the vertical position depends on the level of activity B_α. Assuming the observed spectrum to show a transition to the $1/E_\upsilon^2$ region at $E_\upsilon \approx 10$–20 keV, and using $R_\alpha/E_\alpha \approx 7$ μm/MeV, Eq. (10) gives $d \approx 0.1$–0.2 μm. This could be smaller, since the energy dependence of the anomalous events is not well determined below 10 keV.

The data of Fig. 2 correspond to an NaI crystal with $M \approx 5$ kg, $S \approx 700$ cm^2, $\rho = 3.67$ g cm^{-3}. Substituting these values into (8) together with the observed low energy differential rate $\mathrm{d}n/\mathrm{d}E \approx 0.2$ events/kg/keV/day, the activity level required in the layer is found to be $B_\alpha \approx 10^{-2}\alpha$ g^{-1} s^{-1}.

For a concentration 1 ppb (10^{-9} g/g) of U and Th, the alpha activities for decay chains in equilibrium are

$B_\alpha(\mathrm{U}) = 1.0 \times 10^{-4}\alpha$ g^{-1} s^{-1}.
$B_\alpha(\mathrm{Th}) = 2.4 \times 10^{-5}\alpha$ g^{-1} s^{-1}.

Hence to achieve $B_\alpha \approx 10^{-2}$ one would need either 100 ppb U or 400 ppb Th in the 0.1 μm surface layer. A similar spectrum of anomalous events was seen also in a 2 kg crystal, with a similar required U/Th concentration level.

These levels are three orders of magnitude higher than the intrinsic activity of the NaI crystal (which gives rise to the MeV alpha spectrum in Fig. 2) and would therefore need to have arisen as an independent contamination during crystal manufacture. Because this would be an inexplicably high level of U/Th contamination, we consider now an alternative source of alpha emission, based on radon decay products.

4. Radon decay model

Atmospheric ^{222}Rn transforms to ^{206}Pb via the following principal decay chain:

(i) $^{222}\mathrm{Rn} \rightarrow {}^{218}\mathrm{Po} + \alpha$ ($\tau = 5.5$ d, $E_\alpha = 5.5$ MeV)
(ii) $^{218}\mathrm{Po} \rightarrow {}^{214}\mathrm{Pb} + \alpha$ ($\tau = 4.5$ m, $E_\alpha = 6.0$ MeV)
(iii) $^{214}\mathrm{Pb} \rightarrow {}^{214}\mathrm{Bi} + \beta + \nu$ ($\tau = 39$ m)
(iv) $^{214}\mathrm{Bi} \rightarrow {}^{214}\mathrm{Po} + \beta + \nu$ ($\tau = 29$ m)
(v) $^{214}\mathrm{Po} \rightarrow {}^{210}\mathrm{Pb} + \alpha$ ($\tau = 0.25$ ms, $E_\alpha = 7.7$ MeV)
(vi) $^{210}\mathrm{Pb} \rightarrow {}^{210}\mathrm{Bi} + \beta + \nu$ ($\tau = 32$ y)
(vii) $^{210}\mathrm{Bi} \rightarrow {}^{210}\mathrm{Po} + \beta + \nu$ ($\tau = 7.2$ d)
(viii) $^{210}\mathrm{Po} \rightarrow {}^{206}\mathrm{Pb} + \alpha$ ($\tau = 199$ d, $E_\alpha = 5.3$ MeV)

Reaction (i) deposits ^{218}Po on all available surfaces, cascading to ^{210}Pb which then becomes a long-term source of alpha emission through the 22 year half life of reaction (vi). The key feature of this decay chain is the emission of three alphas in reaching ^{210}Pb via reactions (i), (ii), (v). Each results in a nuclear recoil ~ 100–150 keV which would progressively embed some of the daughter nuclei into the crystal surface, to give an alpha-emitting layer of finite thickness below the surface, as required by the above model.

Each daughter nucleus of mass M_d recoils with energy $E_\mathrm{r} = (M_\alpha/M_\mathrm{d})\, E_\alpha$, giving the following

recoil energies and projected ranges in NaI (calculated from LSS model [10]):

Reaction (i) $E_r = 101$ keV, range $R_1 = 0.039$ μm.

Reaction (ii) $E_r = 112$ keV, range $R_2 = 0.043$ μm.

Reaction (v) $E_r = 146$ keV, range $R_3 = 0.052$ μm.

The 'auto-implantation' distribution can be estimated by performing the definite integrals over z for each of the implantation processes, giving the following results:

In the absence of straggling, a number N isotropically-emitted ^{222}Rn decays on the crystal surface would produce a uniform distribution of ^{218}Po nuclei throughout a layer of thickness R_1:

$$dN/dz = N/2R_1$$

These in turn would produce a distribution of ^{214}Pb (with $R_1 < R_2$):

$$dN/dz = N/4R_2, \quad 0 \leqslant z \leqslant R_2,$$
$$= (N/4R_1R_2)(R_1 + R_2 - z),$$
$$R_2 \leqslant z \leqslant R_1 + R_2.$$

The two beta decays to ^{214}Po produce no significant recoil. The subsequent alpha decay would produce a distribution of ^{210}Pb (with $R_2 < R_3$):

$$dN/dz = (N/16R_1R_2R_3)\left[2R_1R_2 + (z - R_2 + R_3) \times (2R_1 + R_2 - R_3 - z)\right],$$
$$0 \leqslant z \leqslant (R_1 + R_2 - R_3),$$
$$= (N/16R_2R_3)(R_1 + 2R_2)$$
$$(R_1 + R_2 - R_3) \leqslant z \leqslant R_3,$$
$$= (N/16R_2R_3)\left[R_1 + 2(R_2 + R_3 - z)\right].$$
$$R_3 \leqslant z \leqslant R_2 + R_3,$$
$$= (N/16R_1R_2R_3)\left[R_1 + R_2 + R_3 - z\right]^2,$$
$$R_2 + R_3 \leqslant z \leqslant R_1 + R_2 + R_3.$$

The resulting distribution is shown in Fig. 5. Small modifications would result from straggling (~0.02 μm) and from implantation from ^{222}Rn decays a finite distance outside the surface. This distribution can be used to define a z-dependent $B(z)$ in Eq. (7), which after numerical integration results in curve (b) of Fig. 4. It is of interest to note that in this case there is no free parameter to adjust the horizontal position of the spectrum, since the known energies and ranges of the decay alphas have been used. It is therefore remarkable that the position of the $1/E^2$ region is consistent with that of the spectrum of unidentified events in Fig. 2. This results from the fact that the nuclear recoils have a total range which is comparable to the required contamination thickness ~0.1 μm estimated in Section 3 above.

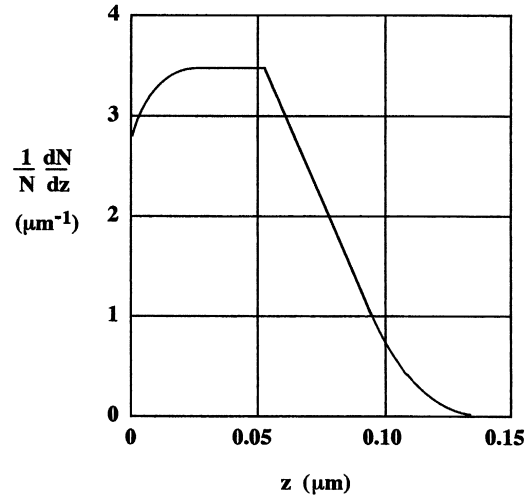

Fig. 5. Approximate activity versus depth profile resulting from recoil nucleus implantation from alpha decays in ^{222}Rn decay chain.

We note that the ^{210}Po concentration which produces the relevant alpha decays via reaction (viii) above would require a period of time to build up from ^{210}Pb decays, the number of Po nuclei being given approximately by (omitting terms from the short ^{210}Bi beta decay):

$$N_{Po} \approx N_{Pb}[\tau_{Po}/\tau_{Pb}] \times \left[\exp(-t/\tau_{Pb}) - \exp(-t/\tau_{Po})\right]. \quad (13)$$

Since $\tau_{Pb} \gg \tau_{Po}$ the build-up time constant is approximately equal to the 200 day decay constant for ^{210}Po. This, however, is shorter than the age of either of the NaI crystals for which the unidentified spec-

trum (Fig. 1) has been observed, so that the ^{210}Po concentration would have been fully established in these crystals.

5. Magnitude of required radon contamination

Curve (b) of Fig. 4 is approximately matched to the observed spectrum by assuming an average $B_\alpha \approx 0.007$ g^{-1} s^{-1} over the implantation depth ≈ 0.12 μm, representing a mass 0.03 g of the 5 kg crystal. Since the time constant of the ^{210}Pb decay is $\tau \approx 32$ y $\approx 10^9$ s, it requires a total implantation of $\sim 2 \times 10^5$ ^{210}Pb nuclei to produce the required level of activity in this layer. The integral of the depth profile in Fig. 5 is ~ 0.3, showing that the recoil process implants 30% of the original surface Rn decays. It thus requires the crystal to have been contaminated by an initial $7c \times 10^5$ Rn decays near the surface.

For a typical atmospheric Rn level ~ 10 decays m^{-3} s^{-1}, depositing Po on all available surfaces, we estimate that even for an exposed crystal the 700 cm^2 surface would receive no more than 0.1–0.5 Po nuclei per second. Thus, if normal air is assumed to be the Rn source, an exposure ~ 1000 h would appear to have been necessary to produce the required ^{210}Pb level. In the case of NaI crystals, this appears to be precluded by the fact that their hygroscopic nature requires isolation in a dry inert atmosphere after cutting from the crystal boule and before subsequent encapsulation, so exposure to the atmosphere should be very short if all procedures are followed correctly. Much longer periods (several years underground operation) are available for diffusion of radon into the detector, but, with the detector sealed inside a ptfe and copper enclosure, with silica windows, a defective seal would effectively expose only a small fraction of the surface area, increasing the above required exposure time by a further 2–3 orders of magnitude.

It remains a possibility that the adjacent encapsulating materials – in particular the ptfe wrapping and quartz windows – could have been exposed for a longer period and not adequately cleaned prior to assembly. For this reason it is important to establish whether the anomalous pulses exist at the same rate in unencapsulated and newly polished crystals.

6. Conclusions

We have shown that it is possible to account both for the existence of a population of low energy alpha events and its approximate spectrum shape, in terms of alpha particles emitted out of the crystal surface from a layer within the crystal of thickness ~ 0.1 μm, very much less than the alpha particle range. To match the observed event rate for unidentified short pulses in NaI, the alpha activity in that layer would need to be ~ 0.01 α s^{-1} g^{-1}, much higher than that of the average intrinsic U/Th contamination and higher than expected from atmospheric Rn contamination.

At least two experiments to test the model could be considered:

(a) A crystal showing the anomalous events could be repolished, removing 0.1μm of surface and retested, preferably in dry nitrogen or vacuum without encapsulation.

(b) An NaI crystal could be exposed to an artificially high radon atmosphere, subsequently observing the predicted build-up of a low energy alpha spectrum from ^{210}Po, fed by ^{210}Pb in accordance with Eq. (13).

Acknowledgements

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