Our short experience at IAS and within ROSEBUD with radioactive contaminations in scintillating bolometers: uses and needs

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Internal radioactive contamination in scintillating bolometers aiming to detect dark matter, which should be absolutely avoided in the ultimate stage of experiments, is a very valuable tool in their definition stage. The goal of this presentation is to report on our past experiences with scintillating bolometers, a mixed "heat and light" detection technique, both at sea level and underground. Focus is given to the last materials tested within the ROSEBUD collaboration in 2007: sapphire, BGO and LiF. An original use of delayed coincidences in the decays from the natural radioactive chains is also presented with the example of a SrF_2 crystal: it highlights position dependence in the light signal which worsens the resolution of this channel.

1. Introduction

The ROSEBUD collaboration is a joint effort between the IAS at Orsay (France) and the University of Zaragoza (Spain) to develop cryogenic detectors able to detect the hypothetical dark matter particles. The experiment is operated underground at the Laboratorio Subterráneo de Canfranc (LSC) but most of the developments and characterization of the prototypes are made at sea level in Orsay, for convenience, as well as for economical reasons: a unique, light weight dilution refrigerator is shared between the two sites. This requires relatively fast detectors with time constants less than some 10 ms in order not to blind the detectors with the cosmic rays at surface.

Scintillating bolometers with typical masses of ~50 g are able to efficiently discriminate between alphas, gammas and nuclear recoils above some tens of keV, at 20 mK, the base temperature of the refrigerator. Particles are discriminated through their ionization power. The technique uses the information provided by both heat and scintillation signals in the target, the latter being detected by an auxiliary light absorbing bolometer, made from a thin disk of Ge and optically coupled to the heat detector in a light reflecting cavity (see Fig. 1 for details on the double bolometer configuration).

The power of the technique relies on the high energy resolution power of the heat channel, found usually in every "good" single bolometer (below 2%), together with an independent measurement of the light emission, signing the nature of the incident particle. In order to explore this new technique, importance has been given first to the test of different materials (from known 300 K scintillators – as BGO, $CaWO_4$,… – to materials known to have excellent thermal properties at low temperature – as sapphire – independently of their radioactive content. As a result, rather high radioactive levels were encountered in the materials tested so far; however, associated events were used to gain a deeper comprehension of the detectors. Some intrinsic contaminations, as ^{207}Bi in BGO, should be reduced for the next generation of detectors.

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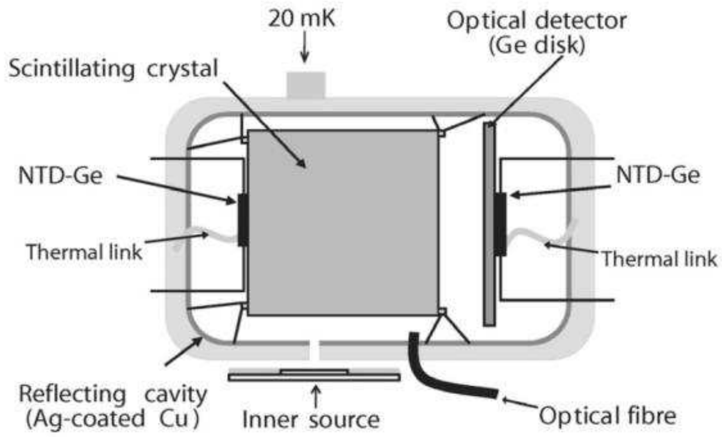

Fig. 1. Schematic view of the double bolometer configuration. Neutron Transmutation Doped Germanium (NTD-Ge) thermistors are used to read both signals (light and heat).

2. The ROSEBUD run in 2007: a complementary set of scintillating bolometers

Scintillating bolometers offer a wide choice of targets. This property is very welcome to face the uncertainties associated with the dark matter particles which are looked for, as its ability to couple to nuclear spin or, if not, the scaling of the cross section with the nuclear content. A set of three double bolometers with targets (from top to bottom) made of 46 g BGO ($Bi_4Ge_3O_{12}$), 33 g LiF and 50 g sapphire (Al_2O_3), each optically coupled to its own optical Ge bolometer, was mounted under the 20 mK mixing chamber of the refrigerator. A complete characterization – thermal responsivities, light yields, discrimination powers – was performed at Orsay before going underground.

Four underground runs, each lasting two weeks, were undertaken in 2007 in the ROSEBUD installation at LSC. Shielding was improved from the February to the May run (increase of lead shielding and removal of radon by nitrogen flushing) reducing the background as can be seen in Fig. 2 and 3. The last run was dedicated to neutron calibration with an external ^{252}Cf source. The results obtained have been analyzed [1, 2], but a complete interpretation of the non-scintillating events seen in the so-called "recoil branch" in the light versus heat discrimination plots is still underway: it will probably need complementary measurements.

A different task was assigned to each bolometer in the experiment: the sapphire one (a low Z material with exceptional thermal properties at low temperature) was recording the low energy events at the keV level, the BGO detector (having 66% Bismuth content in weight) tracked the gamma background profiting from a high efficiency, while the LiF detector attempted to detect the residual neutrons using the ^{6}Li neutron capture reaction. The questions concerning the suitability of these detectors for dark matter detection are only discussed here in the context of their internal radioactive contamination and of analysis of external backgrounds, which is the main concern of this RPScint'2008 workshop. A previous campaign in 2000 allowed us to quantify the radioactivity content of a 54 g $CaWO_4$ detector, which was found to be strongly contaminated in the U-Th chains [3].

2.1. Radio-purity in sapphire

Sapphire is a material with one of the highest melting temperature (~2050 °C). We might expect an important segregation of impurities during the growth of the crystal. The sample used in ROSEBUD was grown from the melt according to the Kyropoulos technique, "somewhere in Russia". No line is seen in the sapphire background within the small exposure time, at low energy (E < 200 keV).

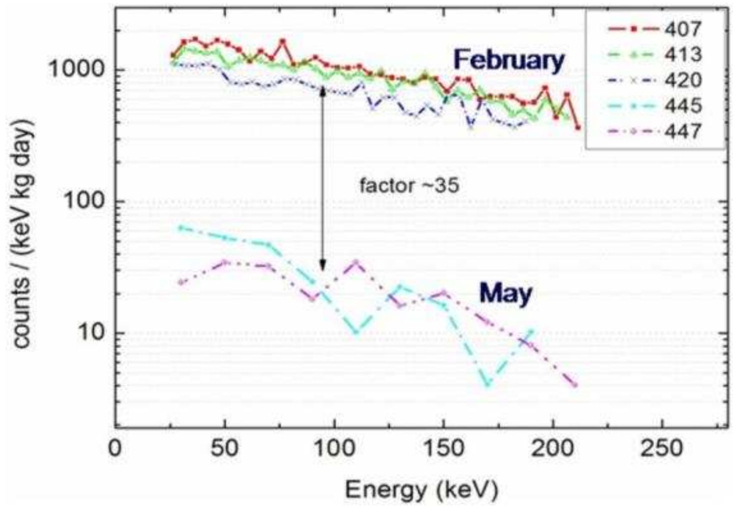

Fig. 2. Improvement of the gamma background in the 50 g sapphire detector at Canfranc between February and May 2007.

Little can be said at higher energy because of the dynamics chosen in the acquisition but one should remind that sapphire is a material difficult to calibrate with high energy gammas. Analysis of internal alpha contamination in the sapphire itself was not addressed in the particular detector tested within ROSEBUD in 2007, which was known to be unintentionally contaminated after a long exposure to a ^{236}Pu source.

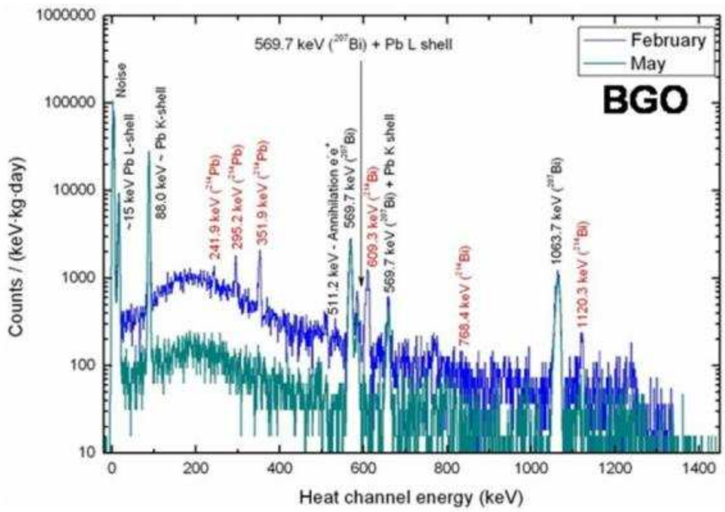

Fig. 3. Improvement of the gamma background in the 46 g BGO detector at Canfranc between February and May 2007. Radon lines disappear in May thanks to nitrogen flushing through the set-up. The lines from EC in ^{207}Bi are dominating this spectrum, with a special mention to the 88 keV line (X-ray and Auger electrons cascade following capture in the ^{207}Pb K-shell).

2.2. Radio-purity in BGO

The 46 g BGO detector presented a 6 keV energy threshold. It was known to be heavily contaminated by ^{207}Bi from previous measurements at Orsay [4]. Using the event rate on the 88 keV line and published branching ratios the internal contamination in ^{207}Bi was estimated at a level of 3300 mBq/kg of BGO (i.e. 5000 mBq/kg of the original bismuth material).

Special attention has been given to the 1063.7 keV gamma events seen in the BGO gamma background spectrum. The 1633.3 keV excited state of ^{207}Pb that feeds this line has a relatively long lifetime ($T_{½}$~0.81 s) while the X rays and Auger electrons cascade emitted when filling the vacancies created by capture in the K, L, M… shells is fully absorbed. The delayed coincidence (cascade + gamma) could be identified in the data and will be used to improve the accuracy of the K/(L+M+…) EC ratios with respect to published ones.

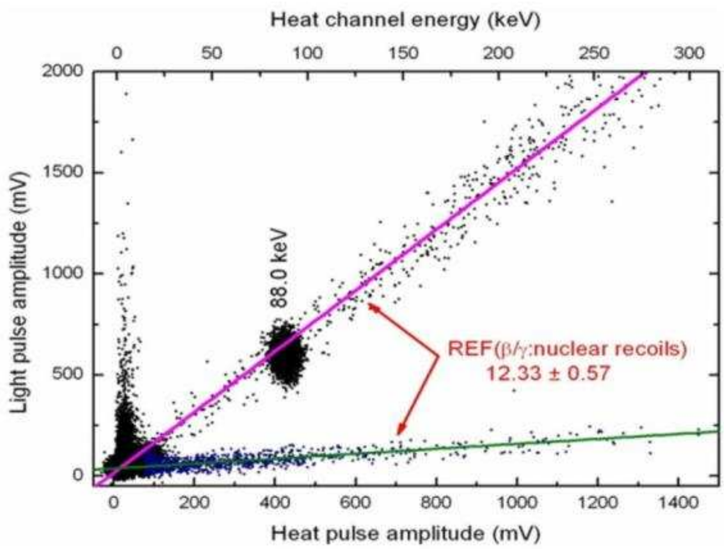

Fig. 4. Light versus heat discrimination plot in the 46 g BGO detector under a ^{252}Cf neutron irradiation. The 88 keV line following EC at the ^{207}Pb K-shell dominates and is well separated from the nuclear recoil events while the 15 keV lines (L-shell) could limit the detector discrimination threshold.

It is however obvious from the “heat and light” discrimination plots registered (see Fig. 4) that such a high contamination level in ^{207}Bi could limit the use of these detectors for dark matter research. As we were aware of this serious drawback of BGO detectors, we made some years ago a compilation of ^{207}Bi content in bismuth containing materials.

Table 1. ^{207}Bi in bismuth compounds.

Source	Activity mBq/kg Bi	Reference and comments
BGO Crismatec (radio-pure quality)	7 ± 2	LSC (1999); J. Puimedón, *private communication*
	9 ± 3	LSM (1999); C. Goldbach (also quoted in mBq/kg BGO: ^{40}K<7; ^{212}Pb=6±4; ^{208}Tl=3±2), *private communication*
BGO Harshaw	175	LSM (<1996?); P. Hubert, *private communication*
BGO Crismatec	750	
PbS concentrate	2000	K. Fukuda et al. (1995) [5]
BGO Crismatec	4400	Y. Satoh et al. (1993) [6]
Bi ingot (1992)	10000	
Bi ingot (1992)	2900	
Bi ingot (1992)	2000	
Bi ingot (1967)	<200	

The BGO detector tested by ROSEBUD in 2007 has a ^{207}Bi content similar to the one tested by Satoh et al in 1993: both crystals were bought from the same company. The origin of such high level of ^{207}Bi in most material tested can be understood if we recall that Bismuth is mostly extracted as a byproduct in Pb metallurgy: the reactions ^{207}Pb(p,n) and ^{206}Pb(p,γ) may occur in lead with protons from the cosmic rays. A second source of ^{207}Bi is anthropogenic: it has been detected in sediments in association with nuclear tests in the 1960-1970's. In principle it could be produced as well in bismuth ores through the ^{209}Bi(n,3n) but fast neutrons are needed. The radio-pure quality BGO developed by the Crismatec company comes, probably, from selected bismuth materials extracted in leadless environments. New detectors were mounted with crystals issued from this radio-pure quality BGO, and they have been tested at sea level in Orsay. The background spectrum was published some years ago [4] and shows lines hardly seen above the continuous background in the heat channel that were attributed to ^{207}Bi, giving a reduction factor in ^{207}Bi content better than 15. However, they are probably due to ^{214}Bi (see D. Grigoriev et al., in these Proceedings) suggesting that the reduction factor is much closer to the expected one (~500-1000).

2.3. Radio-purity in LiF

The scintillating LiF detector aimed to detect environmental neutrons using the neutron capture reaction on ^{6}Li (n+^{6}Li→α+t; Q=4.78 MeV) as shown in Fig. 5. Alpha contaminations in LiF bolometers may be a relevant background source for the estimate of the neutron flux and a light yield improvement would be highly desirable for a better discrimination.

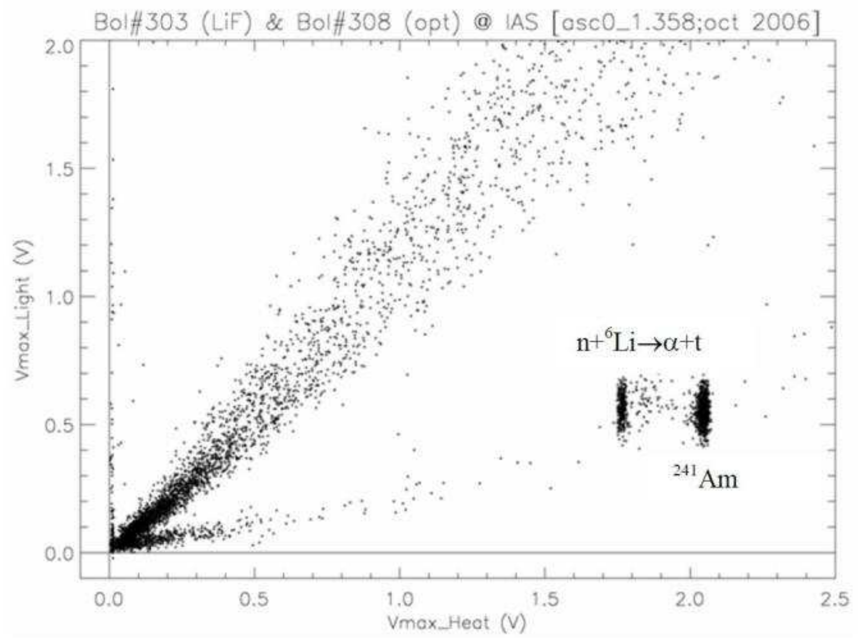

Fig. 5. Light versus heat discrimination plot during a background night at IAS (sea level) as recorded by the 33 g natural LiF detector. A ^{241}Am source has been included in the set-up. While it is obvious that an alpha emits less light than an alpha + tritium pair releasing the same energy, a light yield improvement would be highly desirable for a better separation. Thermal and fast neutrons from the ambient background are detected: the latter detection underlines the need to perform these developments underground to avoid the nuclear recoils following fast neutron scattering in the bolometer targets.

Within the short exposure time during the 2007 runs at LSC, we could hardly detect any significant alpha contamination (see Fig. 6). A calibration with thermal neutrons from a ^{252}Cf source suggests a slight internal ^{210}Po contamination (~mBq/kg) at the level of one count per night, identified at 5.4 MeV. One should recall that bolometers, thanks to their high energy resolution power, can discriminate between internal and external contamination from alpha emitters: the 33 g natural LiF target used at Canfranc showed a better than 40 keV FWHM energy resolution, which is sufficient to resolve external decays (alpha only) from internal ones (alpha + recoil) that are separated ~100 keV.

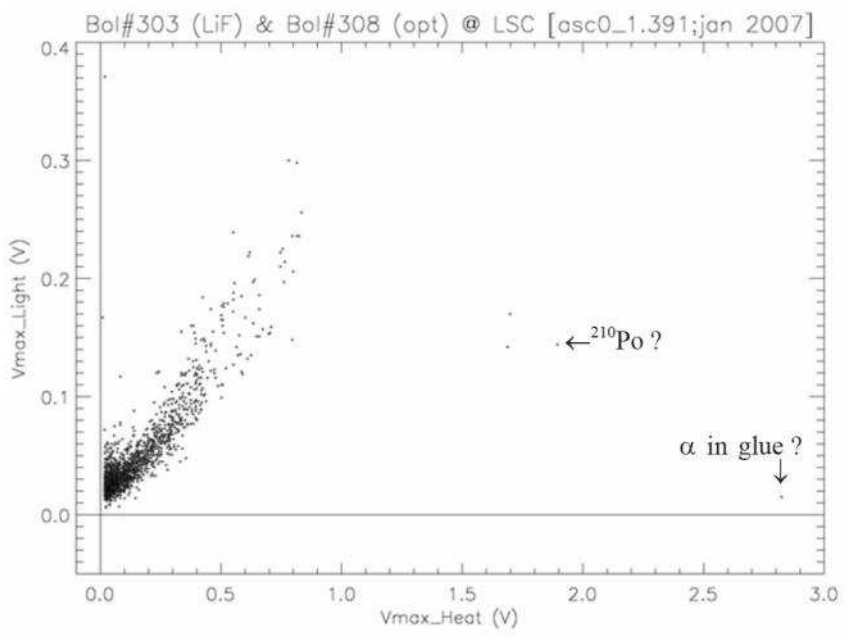

Fig. 6. Light versus heat discrimination plot during a background night at LSC (underground) as recorded by the 33 g natural LiF detector. With respect to the previous figure, the ^{241}Am source has been removed and the strong suppression of the cosmic rays underground can be noticed. The slight ^{210}Po alpha contamination suspected, as well as the single event detected at high energy in a non scintillating part of the detector, which might be attributed to an alpha decay in the glue, are also shown.

A flux of (2-5)$\times10^{-6}$ n/(cm^2 s), in the range of published levels of neutron fluxes underground, has been derived for thermal neutrons inside the low background shielding at LSC. To increase the neutron detection efficiency (both for thermal and fast neutrons) enrichment in ^{6}Li – natural abundance of ^{6}Li is 7% – and/or increasing the mass of the LiF crystal are the solutions proposed for the next step of ROSEBUD in view of EURECA.

3. Use of radioactivity in scintillating bolometers to study the origin of the light signal dispersion. The case for a 54 g SrF_2 crystal

The energy dispersion in the light channel constrains the discrimination efficiency between gammas and nuclear recoils at low energy. It is therefore of the utmost importance to study its origin, but few practical tools are available for this purpose. In particular, one would wish to disentangle light yield inhomogeneities or other geometrical effects from statistical fluctuations.

Cascading alpha decays from the natural radioactive chains could provide such tools. We can illustrate this idea with data taken from a 54 g SrF_2 scintillating bolometer which was found to be highly contaminated in the natural radioactive chains (see Fig. 7). In alpha decays the recoiling nuclei have ranges of about 100 Å. Thus, in a cascading pair, the light emission from both decays is issued virtually from the same point in the crystal (typically cm-sized).

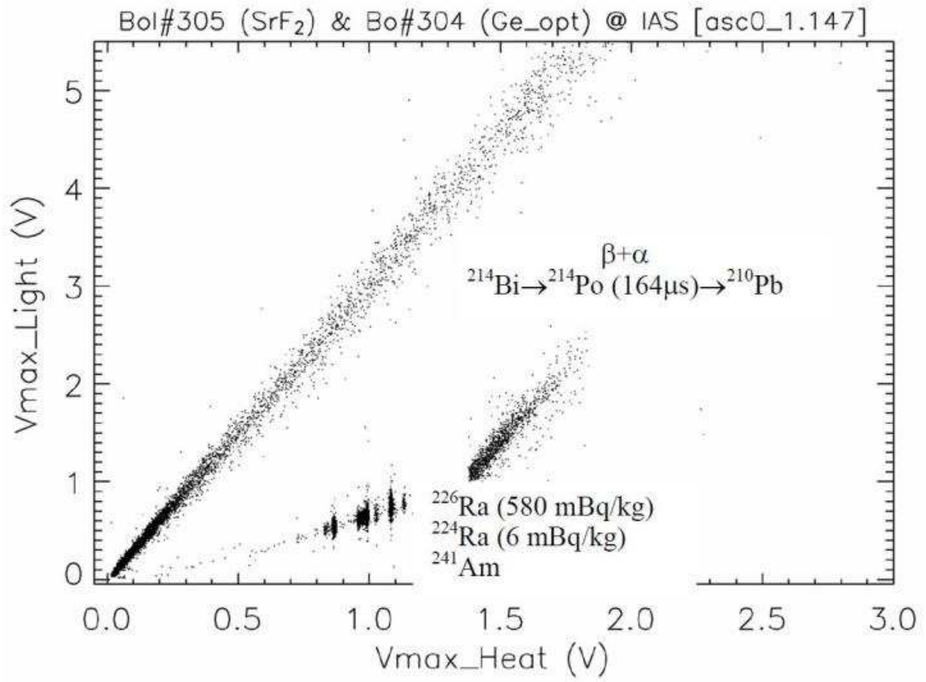

Fig. 7. Background in a 54 g SrF_2 bolometer at IAS which evidences the presence of a high contamination in the natural radioactive chains. A ^{241}Am source was added for calibration purposes.

We tracked the events associated with the following decay cascades:
– ^{224}Ra→^{220}Rn (Q_α=5789 keV; $T_{½}$=3.7 d)
– ^{220}Rn→^{216}Po (Q_α=6405 keV; $T_{½}$=55 s)
– ^{216}Po→^{212}Pb (Q_α=6907 keV; $T_{½}$=150 ms)
which were easily identified, the decay constant of ^{220}Rn being much lower than the mean rate of alpha decays seen in the detector. All decays proceed at 100% with alpha emission. Note that the last decay occurred very often in the same track as the second one, due to the 80 ms recording length chosen in the acquisition. A preliminary analysis of these data is summarized in Fig. 8.

This indicates the existence of a position dependence of the light signal that can be attributed to geometrical origin or inhomogeneities in the crystal. Alphas coming from an external ^{241}Am facing at the detector through a collimated hole show a better energy resolution in the light channel (see Fig. 9) than those measured from the internal contaminations, supporting the above mentioned interpretation.

4. Conclusions

Radioactive contaminations in scintillating bolometers are very useful tools to fully characterize the detectors at a first development step. A complementary target approach, in the spirit of the future EURECA project has been initiated within the ROSEBUD project and was very rewarding. Bigger and ^{6}Li enriched LiF detectors are clearly needed to monitor the neutron flux in a future cryogenic dark matter search experiment like EURECA. Commercial BGO targets suffer from ^{207}Bi contamination at high level, but radio-pure raw material exists with a ^{207}Bi contamination much reduced. A 91g BGO detector made from such target will be studied at LSM in 2009 within the EDELWEISS II installation.

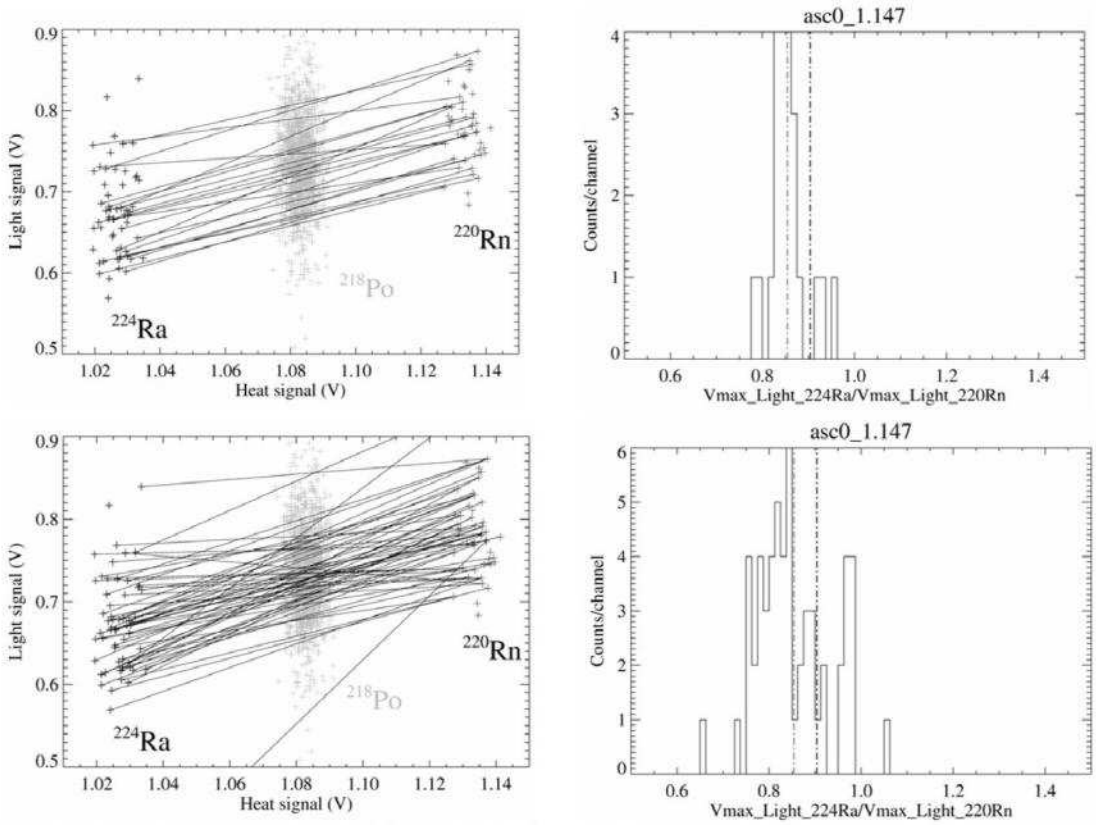

Fig. 8. Light dispersion analysis of decaying pairs in the 54 g SrF_2 bolometer.
Top: Associated pairs from the $^{224}Ra \rightarrow ^{220}Rn \rightarrow ^{216}Po$ are joined by lines. Most of these lines are parallel which suggests a strong correlation of the light emission with the locus of the decay (left). The distribution of the ratios of light signals issued during paired decaying events (dash-dotted line) is slightly shifted with respect to the expected ratio (~0.904), which merely reflects the increasing ionization yield of alphas with energy (right).
Bottom: Artificial, unphysical pairs are created by taking $^{220}Rn \rightarrow ^{216}Po$ decays and the following $^{224}Ra \rightarrow ^{220}Rn$ one, in a kind of time reversal (left). The resulting distribution of the light ratios is much more dispersed.

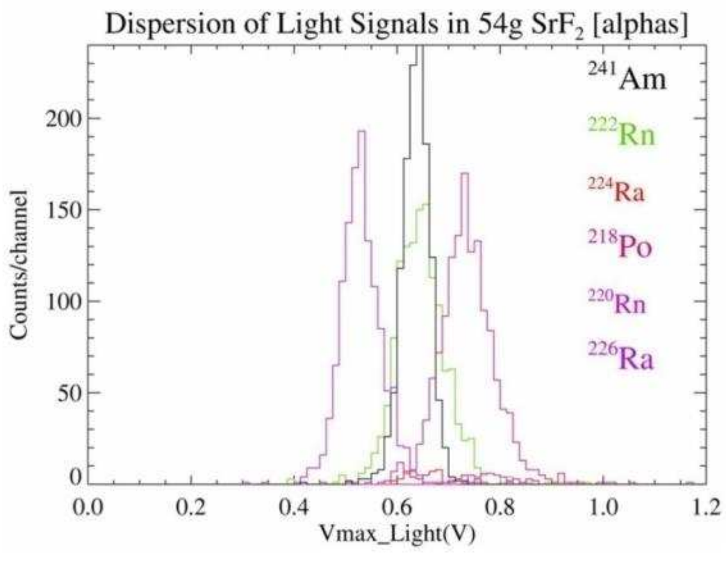

Fig. 9. Dispersion of light signals associated to alpha decays in the 54 g SrF_2 detector.

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