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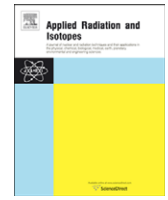

A new low-level γ-ray spectrometry system for environmental radioactivity at the underground laboratory *Felsenkeller*

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A R T I C L E I N F O

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A B S T R A C T

A low-level γ-ray spectrometry system, based on an HPGe-detector with 92% relative efficiency recently installed in the underground laboratory *Felsenkeller* at 110 m water equivalent (w.e.) depth, is described. The integral background count rate normalised to the Ge-crystal mass in the energy range from 40 keV until 2.7 MeV of $0.034\,s^{-1}\,kg^{-1}$ has been achieved by careful material selection of the detector construction material, a graded shielding construction and effective radon suppression. The detector is highly suitable for the effective surveillance of water for human consumption with decision thresholds for ^{226}Ra and ^{228}Ra in the order of some $mBq\,L^{-1}$.

1. Introduction

The radionuclides ^{226}Ra and ^{228}Ra are among the most significant natural radionuclides for the radiological exposition from ingestion of tap or bottled water. Therefore the analysis of these radionuclides has recently gained in importance. If, for example, bottled water should be suitable for the consumption of children below one year, the German legislation calls for maximum activity concentrations for ^{226}Ra and ^{228}Ra of 125 and $20\,mBq\,L^{-1}$, respectively (Min/TafelWV, 2006). For tap water instead of activity concentration limits an annual ingestion dose limit of 0.1 mSv is specified (Council Directive 98/83/EC, 1998). This parameter has to be calculated using nuclide specific activity concentrations, age related consumption rates and dose conversion factors. Until now the discussion in Germany is still ongoing how this ingestion dose should be calculated. Under the most restrictive conditions, e.g. if the ingestion dose limit should also be fulfilled by the most critical age group, the children below one year, the requirements are as stringent as for bottled water. For a continuous surveillance the laboratory is thus obligated to guarantee decision thresholds of approximately 10% of the mentioned reference values, e.g. 10 and $2\,mBq\,L^{-1}$ for ^{226}Ra and ^{228}Ra, respectively. If both radionuclides have to be analysed simultaneously, γ-ray spectrometry is the most suitable and usually used analytical method.

To reach the required detection limits it is common practice to evaporate large sample volumes (>20 L) to get a volume suitable for direct γ-ray spectrometry in *Marinelli* beaker geometry (volume from 0.45 to 2.5 L). Beside this standard procedure there are other ways of reducing detection limits. The full energy peak efficiency can be increased by using larger detectors and more point like sample geometries, by for example performing $Ba(Ra)SO_4$ precipitation. Background reduction is another way for lowering the decision limits of samples containing only traces of the radionuclides under investigation. The background of low-level γ-ray spectrometry systems located above ground is dominated by the cosmic ray induced continuous background. Installing an active anti-muon veto (Laurec et al., 1996; Schwaiger et al., 2002) or going underground (Reyss et al., 1995; Arpesella, 1996; Niese et al., 1998; Hult et al., 2005; Neumaier et al., 2000; Neder et al., 2000) is the alternative for further continuous background reduction. Underground installations, in the depth region from 10 to 100 m water equivalent (w.e.), often combine additional anti-muon detectors as well (Heusser, 1991; Hentig, 1999; Semkow et al., 2002; Povinec et al., 2004).

Here, a new γ-ray spectrometry system mainly dedicated to the detection of activity concentrations in the order of some $mBq\,L^{-1}$ in tap and bottled water is described. The detector, without anti-muon veto, was installed 2007 in the underground laboratory *Felsenkeller* (Niese et al., 1998) at a depth of 110 m w.e.

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Table 1
Specific activity (in $mBq\,kg^{-1}$) of selected detector construction materials for *D*6 and mass *m* of these materials in the final detector assembly.

Material	m/g	^{238}U	^{226}Ra	^{228}Ra	^{228}Th	^{40}K	^{137}Cs	^{60}Co	Laboratory, detector, sample mass, measurement time
ULB-Al	2256	<15	<0.70	<0.9	3.8±0.7	4.9±1.8	<0.20	–	Gran Sasso, GeMPI, 4.0 kg, 18 d
Cu	1600	<35	<1.2	<1.1	<1.8	< 12	<0.70	<0.70	Gran Sasso, GeMPI, 3.4 kg, 7 d
Isolator 1, batch 1	10	285±100	17±3	79±8	190±10	< 55	<2.4	<1.3	Gran Sasso, GeMPI, 68 g, 32 d
Isolator 1, batch 2	10	<590	<90	<66	<56	< 730	<25	–	Gran Sasso, GePaolo, 10 g, 21 d
Brass screws	20	–	280±20	–	–	900±250	–	–	UDO, ULB, 7.8 g, 14 d
Screws from old ships iron[a]	20	–	0.15±0.04	0.46±0.14	0.46±0.14	1.0±0.4	<30	<18	Gran Sasso, GeMPI, 38.1 kg, 80 d

[a] Heusser et al. (2006).

2. Experimental

2.1. Material selection for the Ge detector

The key part of the new low-level γ-ray spectrometry system (further referenced as *D*6) is a coaxial p-type HPGe-detector of 92% relative efficiency with enhanced front-side sensitivity for the low energy region (*CANBERRA* XtRa® GX 90-205). The germanium crystal has a diameter of 78 mm and a length of 75 mm, which is equivalent to a volume and mass of the active germanium of 362 cm^3 and 1.93 kg, respectively. The FWHM of the detector at 1.332 MeV was determined as 1.9 keV.

All materials used for the construction of the detector were measured for radiopurity within the *CELLAR*-laboratories (*C*ollaboration of *E*uropean *L*ow-*L*evel Underground *LA*bo*R*atories) before assembling them. Analyses were performed on at least 17 different material types for more than 200 d measurement time in total. It turned out that the most crucial materials for the detector background are the ultra low background aluminium (ULB Al), which represents the largest mass in the final assembly and some isolators and brass screws, which may contain enhanced levels of natural radionuclides (Table 1). Careful material examination led to the following consequences in the material selection and changes in constructive details in order to achieve a nearly peak free background spectrum under the conditions of the *Felsenkeller* laboratory:

(a) Screws manufactured from old ship iron were used instead of brass screws near the Ge crystal.
(b) The isolator 1 was replaced with a more radiopure piece of the same material.
(c) Finally, additional 9 mm archaeological lead and 1 mm radiopure copper was positioned inside the endcap between the germanium crystal and the isolator 2 (see Fig. 1).

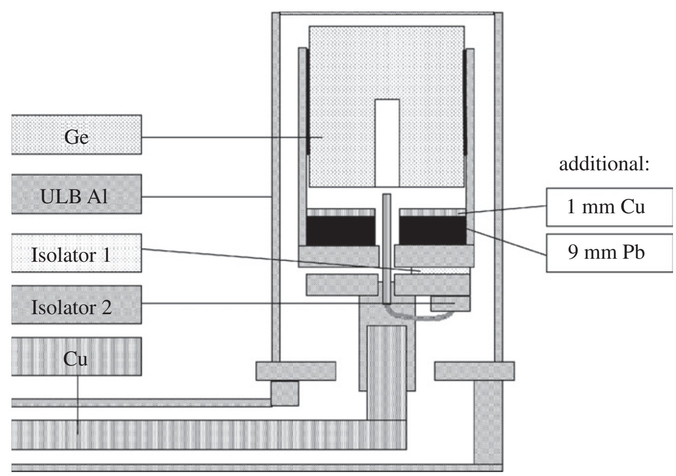

Fig. 1. Schematic view of the endcap of the detector *D*6.

2.2. Description of the passive shielding

The passive shielding (Fig. 2) consists of 5 cm of copper, 5 cm of low-activity lead from *Plombum*, Poland, with (2.7±0.6) $Bq\,kg^{-1}$ of ^{210}Pb and additionally 10 cm lead from *van Gahlen*, Netherland, with (33±4) $Bq\,kg^{-1}$ of ^{210}Pb. In order to reduce the production of neutron induced γ-emitters the OFHC (oxygen-free high conductivity) copper was stored underground immediately after production and brought above ground only for a short processing period of some days. Special effort was spent to reduce the background from the omnipresent gaseous ^{222}Rn, which has a concentration in the laboratory air of (60±20) $Bq\,m^{-3}$. Therefore the space between the detector endcap and the copper layer of the shielding was reduced drastically to a gap of less than 4 mm. Additionally, the whole shielding was gastight housed and flushed with gaseous nitrogen from the detector's Dewar. Beside the possibility to measure cylindrical samples (∅⩽90 mm, height⩽80 mm) on top of the endcap of the detector, rings of the innermost copper layer are removable and allow alternatively positioning of a 2.5 L *Marinelli* beaker. The lid of the shielding with a mass of about 80 kg can be opened with the help of an electrical drive unit.

The γ-ray spectrometry system is positioned in the measuring chamber 2 of the medium deep underground laboratory *Felsenkeller*. The 50 cm thick walls of the chamber with the internal dimensions of $(3\times6\times2.2)\,m^3$ were realised as a sandwich construction of steel and lead with a final area density of 210 $g\,cm^{-2}$. They reduce the photon equivalent dose rate to less than 5 $nSv\,h^{-1}$, whereas in the unshielded cave outside the chamber a dose rate of 450 $nSv\,h^{-1}$ was obtained due to the enhanced levels of primordial radionuclides in the rock (150 $Bq\,kg^{-1}$ ^{238}U, 200 $Bq\,kg^{-1}$ ^{232}Th and 1300 $Bq\,kg^{-1}$ ^{40}K). The muon flux in the laboratory is reduced by a factor of 42 compared to the ground level value. Furthermore the thermal neutron fluence rate was determined with activated Au and Ta targets to be less than 2 $m^{-2}\,s^{-1}$.

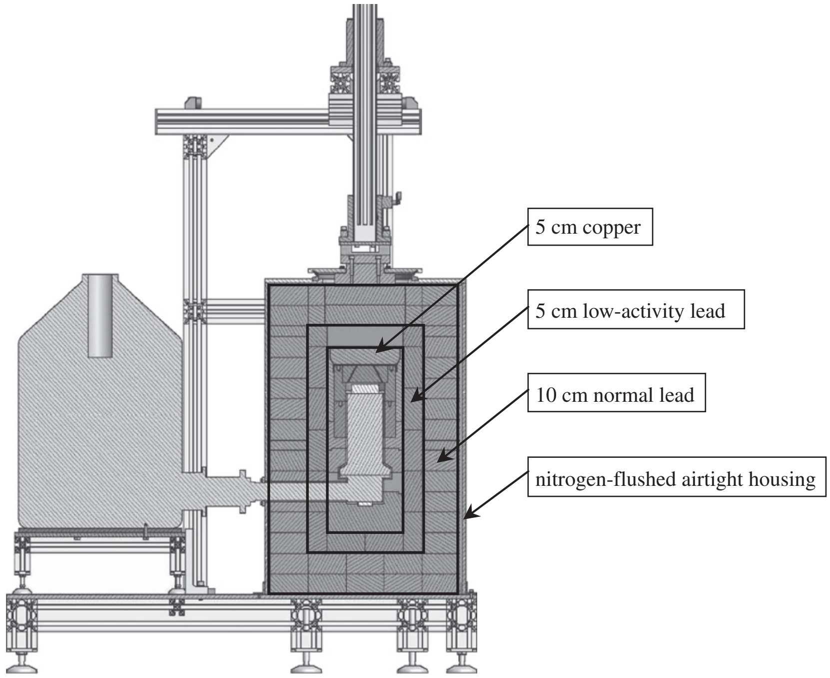

Fig. 2. Schematic view of the shielding of the detector *D*6.

3. Results and discussion

3.1. Background characteristics

The background spectrum of *D*6 (Fig. 3) (58 d measurement time) shows only four peaks. Beside the 511 keV annihilation peak, the clearly visible 1124 keV peak corresponds to the neutron activation of Ge to ^{65}Zn (sum of $E_\gamma = 1115.5$ keV and $E_x = 8.1$ keV) during the ground level handling of the Ge crystal. The obtained count rate of $(2.9 \pm 0.5)\,d^{-1}$ stays in accordance with Heusser (1993) who presented a value of $1\,d^{-1}$ for a 1 kg Ge detector. The small peaks from ^{212}Pb of $(3.2 \pm 0.8)\,d^{-1}$ and ^{40}K of $(1.9 \pm 0.4)\,d^{-1}$ originate most likely from the remaining activity of these radionuclides in the ULB aluminium of the detector's construction material. This residual background implicates no restrictions for typical routine measurements of some days. The obtained background count rates and decision thresholds for the most important primordial and selected artificial radionuclides are expressed in terms of count rates in Table 2. The comparison with the background count rates of other detectors in the laboratory (here with a coaxial n-type detector of 30% relative efficiency, further referenced as *GMX*3) shows a clear improvement by the new system *D*6.

The integral background normalised to the mass of the Ge-crystal in the energy range from 40 until 2.7 MeV was determined as $0.034\,s^{-1}\,kg^{-1}$, a factor of 38 better than an installation in our institution at ground level and a factor of 3.2 better than the older coaxial detector *GMX3* installed also in the *Felsenkeller* laboratory (see Table 3). Due to the only partial muon suppression in the state of the art systems at ground level with anti-muon veto (Laurec et al., 1996; Schwaiger et al., 2002) the integral background count rate there is still a factor of 4.7 higher than the here obtained value without veto at a depth of 110 m w.e. Nevertheless, the obtained improvement using an anti-muon detector 35 m w.e. underground (Povinec et al., 2004) illustrates the reduction potential for the integral background count rate by an additional factor of 3. The comparison with an installation with larger overburden (e.g. Wieslander et al., 2009) shows that at the depth of the *Felsenkeller* laboratory events produced by muon interactions still dominate the detector background.

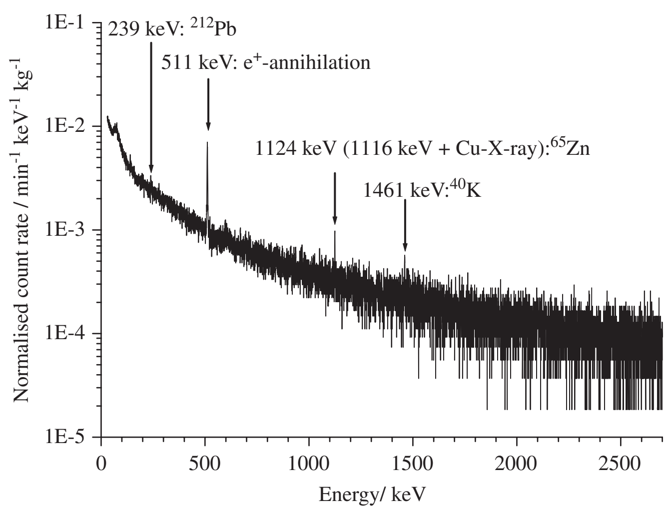

Fig. 3. Background spectrum of the detector *D*6 ($t = 58$ d).

The usage of copper as an inner layer of a graded shielding attenuates the bremsstrahlung continuum and the X-rays induced by the betas from the ^{210}Bi decay ($E_{\beta max} = 1.2$ MeV) in the lead. The usage of Cu will, however, in ground level or medium depth systems increase the continuous background in the region between 100 and 500 keV (Heusser, 1993). On the other hand the usage of copper at deep underground installations is well established (Neder et al., 2000; Neumaier et al., 2000). The question was therefore investigated, if the usage of additional 5 cm Cu as inner layer of a shielding construction in a medium deep underground laboratory (like *Felsenkeller)* will increase the background. The integral count rate in the region from 40 to 500 keV was determined as $(0.0451 \pm 0.003)\,s^{-1}$, whereas after removing of the inner copper of 2 cm thickness a 20% higher count rate of $(0.0547 \pm 0.003)\,s^{-1}$ was obtained. It can therefore be stated that under the conditions of the *Felsenkeller* laboratory the additional usage of copper as inner layer of a 15 cm lead shielding will still decrease the background in the low energy part of the spectrum.

A shielding construction with a minimised cavity, as it is realised here with a gap of only 4 mm between detector and innermost copper, will increase the continuous background during sample measurement by backscattering. However it is shown in Table 4, that the reduction of the background by 2 cm Cu prevails the effect of backscattering as long as the activity of the sample is lower than 2 Bq. Since the system is dedicated to the measurement of samples with activities in the range of some mBq the realised construction with a minimised cavity is reasonable.

Table 2
Comparison of the background count rates R_0 of the 92% HPGe detector *D6* ($t = 58$ d) and of the 30% n-type detector *GMX3* ($t = 13$ d)[a].

E_γ (keV)	Nuclide	R_0 (d^{-1})	
		D6	GMX3
46	^{210}Pb	<2.9	29±3
63	^{234}Th	<2.9	10±3
93	^{234}Th	<2.4	17±3
186	^{226}Ra+^{235}U	<1.7	9±3
239	^{212}Pb	3.2±0.8	<5.4
295	^{214}Pb	<1.5	<4.8
338	^{228}Ra	<1.3	<4.3
352	^{214}Pb	<1.3	4.3±2.1
583	^{208}Tl	<1.0	<2.6
609	^{214}Bi	<1.1	2.8±1.3
662	^{137}Cs	<1.0	8.7±1.4
911	^{228}Ra	<0.84	<2.2
968	^{228}Ra	<0.81	<2.3
1120	^{214}Bi	<0.80	<2.1
1124	^{65}Zn+X	2.9±0.5	<1.8
1173	^{60}Co	<0.71	5.8±1.2
1332	^{60}Co	<0.71	6.2±1.1
1461	^{40}K	1.9±0.4	8.0±1.2
1764	^{214}Bi	<0.63	1.7±0.8
2614	^{208}Tl	<0.51	1.3±0.5

[a] The count rate is reported together with the assigned standard uncertainty. Decision thresholds are given according to DIN 25482-5 (1993) with $\alpha = 0.025$.

3.2. Decision thresholds

The decision threshold g^* expressed as activity for a selected peak energy is given under the assumption of an error probability of the first kind of $\alpha = 0.025$ and the parameters $b = 2l$ as (DIN 25482-5, 1993)

$$g^* = \frac{1.92}{\varepsilon P_\gamma t}\left(1 + \sqrt{1 + 2.08 R_0\, t}\right)$$

where ε is the full energy peak efficiency for the sample-detector geometry, P_γ the emission probability, t the measurement time in s and R_0 the count rate in a region of 2.5 FWHM beside the peak under investigation in s^{-1}.

Fig. 4 shows the energy dependence of the decision threshold g^* under the assumption of $P_\gamma = 1$ and $t = 1$ d for the geometries *filter* ($\varnothing = 47$ mm, height = 1 mm) and *cylinder* ($\varnothing = 47$ mm, height = 6 mm), which where in routine analyses used for the determination of radium isotopes after $BaSO_4$ precipitation. Additionally to detector *D6* the decision thresholds for the older *GMX3* detector are included. It is clearly seen that the decision threshold reaches its minimum at energies of approximately 100 keV, where the full energy peak efficiency is maximum. For the larger detector *D6* the difference between geometries with different sample heights is nearly negligible because the Ge diameter ($\varnothing = 78$ mm) is much larger than the sample one

Table 4
Effect of the innermost Cu layer to the counting rate R in the energy range from 100 keV until 150 keV.

Experimental setup	With ^{60}Co source		Without
	R (min^{-1})	R (min^{-1} Bq^{-1})	R (min^{-1})
Minimised cavity[a]	235	–	0.44
Increased cavity[b]	187	–	0.52
Difference	48	0.040	−0.080

A ^{60}Co source in filter geometry of (1200±120) Bq was used to investigate the influence of backscattering during sample measurements.
[a] 15 cm Pb/5 cm Cu/0.4 cm air/detector.
[b] 15 cm Pb/3 cm Cu/2.4 cm air/detector.

Table 3
Integral background count rates R_{int}, normalized to the mass of the Ge crystal, of above ground, shallow and medium depth γ-ray spectrometry installations with and without anti-muon veto.

Depth (m w.e.)	Location	Detector	Ge mass (kg)	Energy range (keV)	R_{int} (s^{-1} kg^{-1})		Reference
					Without veto	With veto	
0	Dresden, Germany	*D7*	0.85	40–2000	1.3	–	–
0	Daejon, South Korea	–	1.5	50–3000	0.83	0.23	Byun et al. (2003)
0	Seibersdorf, Austria	–	0.94	40–2700	1.1	0.18	Schwaiger et al. (2002)
0	Bruyères-le-Châtel, France	–	1.0	30–2700	0.84	0.16	Laurec et al. (1996)
15	München, Germany	–	3.3	40–2000	0.32	0.074	Hentig (1999)
15	Heidelberg, Germany	–	0.90	65–2680	0.41	0.030	Heusser (1991)
33	Albany, USA	–	2.58	50–2700	0.26	0.13	Semkow et al. (2002)
35	Monaco	1	2.15	40–2700	0.094	0.016	Povinec et al. (2004)
		4	4.18		0.062	0.012	
110	Dresden, Germany	*BL*	0.80	40–2700	0.045	–	This work
		GMX3	0.85		0.11	–	
		D6	1.93		0.034	–	
500	Mol, Belgium	*G4*	2.19	40–2700	0.0028	–	Hult (2008)

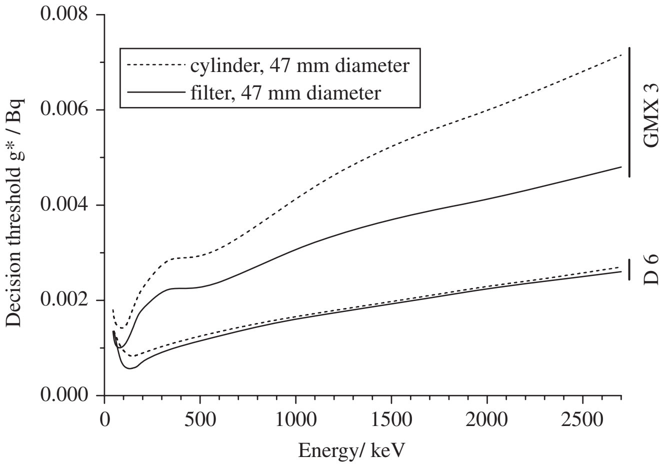

Fig. 4. Decision threshold g^* versus energy for measurement time of $t = 1$ d and normalised emission probability of $P_\gamma = 1$.

Table 5
Comparison of the decision thresholds g^* for ^{226}Ra and ^{228}Ra in *filter* geometry between 92% detector *D*6 and 30% detector *GMX*3 ($t = 1$ d).

Nuclide	E_γ (keV)	g^* (mBq)	
		*D*6	GMX3
^{226}Ra	186	20	49
	295	5.2	12
	351	2.9	6.5
	609	3.0	5.4
	1120	12	23
	1764	14	25
^{228}Ra	338	8.9	21
	911	6.2	11
	968	10	19

($\varnothing = 47$ mm). Only for energies below 200 keV where self-absorption inside the sample has more influence the *filter* geometry has some advantages. For the smaller detector *GMX*3 a larger difference between the two geometries can be obtained, because the Ge diameter ($\varnothing = 53$ mm) is nearly equal to the sample one. Additionally, the curves show a clear bump below 500 keV for *GMX*3. This can be explained by the increased background due to the bremsstrahlung continuum from the lead of lower quality, which was characterised by a specific ^{210}Pb activity of 30 Bq kg^{-1} for *GMX*3 instead of the 2.7 Bq kg^{-1} inside the new installation *D*6. For the mentioned geometry and energies below 400 keV an improvement of the reachable decision threshold for the new detector *D*6 by a factor of larger than 2 can be stated.

Finally, Table 5 presents decision threshold expressed in terms of activities for ^{226}Ra and ^{228}Ra. The required decision thresholds for the surveillance of water for human consumption can be reached for a measurement time of 1 d in the geometry *filter* after $Ba(Ra)SO_4$ precipitation of a 5 L sample volume.

4. Conclusions

The described low-level γ-ray spectrometry system is well suited for the conditions in the *Felsenkeller* laboratory at 110 m w.e. The integral background normalised to the Ge-crystal mass in the energy range from 40 keV until 2.7 MeV was determined as 0.034 s^{-1} kg^{-1}. This has been achieved by careful material selection of the detector's construction material, a graded shielding construction and effective radon suppression. The detector *D*6 provides an ideal tool for the effective surveillance of water for the human consumption with decision thresholds for ^{226}Ra and ^{228}Ra in the order of some mBq L^{-1}.

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