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Section A

The suppression of beam-related background in the ILL neutron–antineutron oscillation experiment

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The search for neutron–antineutron ($n\bar{n}$) oscillations conducted by a Heidelberg–ILL–Padova–Pavia collaboration at the European slow neutron facility at Grenoble requires a very strong cold neutron beam, but an extremely low neutron-induced radiation background. This article describes the analysis and suppression of neutron-beam-related background in the $n\bar{n}$ experiment. In particular, the fabrication of a 20 m^2 shield of isotopically pure sintered ^{6}LiF ceramics which protects the annihilation detector from the inside against neutrons scattered from the annihilation target is described.

1. Introduction

A search for neutron–antineutron oscillations was conducted at the European slow neutron facility of the Institut Laue–Langevin (ILL) at Grenoble [1]. In this experiment a cold neutron beam of intensity $\Phi_n = 1.3 \times 10^{11}$ s^{-1} passes a thin carbon foil of 1.1 m diameter after a free flight of about 70 m in vacuum. An antineutron created from a neutron in the free-flight region would annihilate in this target into an average of five pions. The annihilation products are detected in the ~ 100 m^3 large annihilation detector which surrounds the target.

This experiment is very sensitive to the radiation background that inevitably accompanies a very strong slow neutron beam. As compared with the first $n\bar{n}$ experiment at ILL [2], the neutron intensity has been increased by two orders of magnitude, whereas the requirements on the beam-related background remained about the same.

Although the neutron beam is clean in the sense that it contains no other particles than cold neutrons, any neutron which is lost near the beam will create prompt capture gamma radiation, and quite often also long-lived radioisotopes. The beam-related background in the detector region should be below some 10^6 gamma rays per second. Hence with more than 10^{11} neutrons per second passing through the target, less than 10^{-5} of all neutrons are allowed to contribute to detector background.

This is a formidable problem because a slow neutron beam resembles more a gas jet than a well-defined particle beam. Free neutron beams typically have rather large diameters (up to 1 m^2 in our case) and a rather large angular spread (some cm/m). Furthermore, the neutrons have an approximate Maxwellian velocity distribution, and in the long $n\bar{n}$ free-flight tube the slowest neutrons will fall in the earth's gravitational field far enough to hit the inner surface of the 1.4 m diameter neutron beam tube. Finally, slow neutrons

have large scattering cross sections on most materials, and often also considerable radiative capture cross sections. The solutions to the neutron beam tayloring problems in the nn̄ experiment will be discussed elsewhere. Here we describe the measures taken to analyze and suppress the main beam-related detector background, which was due to the neutron interactions with the annihilation target.

We first report some measurements to determine the composition and structure of the high purity carbon foil used as the annihilation target. Then we discuss the composition of the residual radiation background as obtained from a number of separate investigations. Finally, we describe the fabrication and installation of a shield of pure ^{6}LiF ceramics which protects the inner wall of the neutron beam tube inside the detector over a surface of about 20 m^2 from neutrons scattered off the annihilation target, see fig. 1.

Both carbon and ^{6}LiF are basic materials in neutron research. Neutron-induced background from both materials is low and its sources were difficult to determine. We therefore think that this description is of more general interest.

2. Calculated neutron-induced background rates from the target

The annihilation target in the nn̄ experiment should have the following properties:

1) absorptive for cold antineutrons;
2) transparent for cold neutrons;
3) of low atomic mass number Z to prevent pion reabsorption within the target nuclei;
4) mechanically stable at room temperature and in vacuum.

The annihilation cross section for cold antineutrons is large enough for requirement no. 1 to be no problem for any foil of thickness of some tenths of a millimeter. The requirements of low neutron scattering, of extremely low neutron absorption cross sections, and of low Z (nos. 2, 3) are met only by deuterium, beryllium and carbon.

To be mechanically stable (no. 4) deuterium can only be used in the form of deuterated hydrocarbons. These materials usually pose problems of purity and, in particular, of isotopic purity: protonic hydrogen with its large neutron absorption ($\sigma_a = 0.33$ b at neutron velocity $v = 2200$ m s^{-1}) and very large neutron scattering cross section ($\sigma_s \approx 70$ b) is especially unwanted as an impurity. Beryllium, on the other hand, produces no neutron capture gamma rays and would make a very good annihilation target. However, not only are large thin beryllium foils very expensive, but the residual impurities in even the purest available foils (99.8 + %) produce the same number of capture gamma rays as a pure carbon target of the same mass.

Therefore, carbon, with $\sigma_a = 0.0035$ b and $\sigma_s = 4.7$ b was chosen as target material, in the form of exfoliated graphite with the tradename of grafoil, of thickness 0.019 g cm^{-2}, and 99.99% purity. The same target had been used in earlier neutron oscillation experiments [2,5]. From carbon one expects three main capture gamma lines at respectively 4.94 MeV (67%), 3.68 MeV (33%) and 1.26 MeV (32%), with a total gamma intensity of

$$\Phi_\gamma(\mathrm{C}) = 1.3\rho\sigma_a\Phi_n^{\mathrm{capt}} = 2.2\times 10^6\ s^{-1} \quad \text{(calc.)} \qquad (1)$$

with the cold neutron "capture flux" $\Phi_n^{\mathrm{capt}} \approx 4\Phi_n = 5 \times 10^{11}$ s^{-1} and the target atomic density of $\rho = 1.0 \times 10^{21}$ cm^{-2}. This is the unavoidable background compared with which all other noise sources should be small. The gamma ray background expected from target impurities should be negligible, according to the

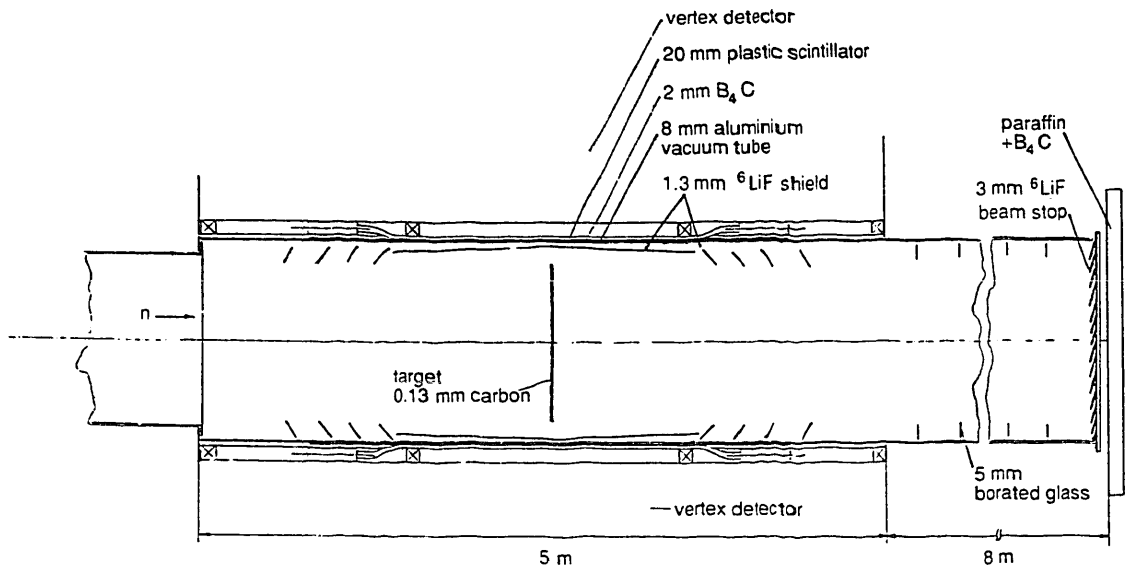

Fig. 1. The target region of the nn̄ experiment and its surroundings.

suppliers information (30 ppm of Fe, 26 ppm of Si, 11 ppm of Na, etc.).

The next problem concerns the number of neutrons scattered off the target. As neutron scattering on carbon, with a cross section of $\sigma_s = 4.7$ b, is all coherent, this number strongly depends on the atomic structure of the material. From a Monte Carlo study based on the measured neutron velocity spectrum and the crystalline structure of grafoil we arrived at a fraction of 5×10^{-3} neutrons scattered off the target, or

$$\Phi_n^{\text{scatt}}(\text{C}) \leq 5 \times 10^{-3} \Phi_n = 7 \times 10^8 \text{ s}^{-1} \quad \text{(calc.)}. \qquad (2)$$

This scattered fraction of 5×10^{-3} may be overestimated: in the calculation it was assumed that the orientations of the basal planes in grafoil have an isotropic distribution; in reality these planes are oriented preferentially parallel to the foil surface, with a 30° FWHM in their angular distribution. This means that first order Bragg scattering is dominant in the beam direction and would in part miss the detector.

In any case the scattered neutron rate (2) is far too high to be tolerable in the nn̄ detector. To intercept the scattered neutrons the inner side of the 1.4 m diameter detector beam tube was covered with tiles of ^{6}LiF ceramics, as described in a later paragraph. The thickness of the tiles was 0.13 cm, or 0.30 g cm^{-2}, and was chosen so as to intercept a sufficient number of neutrons. The tiles should not be much thicker in order to keep the attenuation of pions at a minimum and also to reduce the mass of enriched ^{6}Li needed. The transmission T of this shield depends exponentially on the neutron velocity: $T = 1 \times 10^{-9}$ for $v = 550$ m s^{-1} and $T = 6 \times 10^{-3}$ for $v = 2200$ m s^{-1}. The total transmitted intensity of neutrons reaching the detector was calculated, for a usual cold neutron spectrum, to be $\langle T \rangle = 3 \times 10^{-5}$, or

$$\Phi_n^{\text{trans}}(^6\text{LiF}) = \langle T \rangle \Phi_n^{\text{scatt}} = 2 \times 10^4 \text{ s}^{-1} \quad \text{(calc.)}, \qquad (3)$$

which is negligible compared with (1).

^{6}LiF is one of the few materials that efficiently capture thermal neutrons with only very small secondary particle emission rates. For the ^{6}Li enrichment of 96% used in the ^{6}LiF shield the residual gamma ray intensity is again very small

$$\Phi_\gamma(^6\text{LiF}) = 4 \times 10^{-5} \Phi_n^{\text{scatt}} = 3 \times 10^4 \text{ s}^{-1} \quad \text{(calc.)}, \qquad (4)$$

In one out of 10^4 neutron capture events in ^{6}LiF fast neutrons are produced [3] with energies up to 10 MeV. But the fast neutron flux originating in the ^{6}LiF shield and hitting the nn̄ detector is again rather low:

$$\Phi_{\text{fast n}}(^6\text{LiF}) = 0.7 \times 10^{-4} \Phi_n^{\text{scatt}} = 5 \times 10^4 \text{ s}^{-1} \quad \text{(calc.)}. \qquad (5)$$

3. Measured target properties and background rates

The calculated radiation background rates (1) and (3) to (5) originating from the annihilation target should be tolerable for the nn̄ detector. However, when the neutron beam was opened it was found that the detector background due to the target was much larger than expected from (1) to (5), and was by far the dominant (single rate) background in the trigger detectors. A number of different investigations were necessary to clarify this issue.

First it seemed as if more neutrons were absorbed or scattered off the target than expected from (1) and (2) because a neutron total transmission experiment on a stack of target foils showed transmission losses of about 3%, instead of the calculated 0.5% from (2). A neutron transmission time-of-flight measurement with a chopped beam gave the neutron velocity dependence of the transmission cross section shown in fig. 2, together with the initial neutron velocity spectrum. (Due to a different geometry, this incoming neutron velocity spectrum is not the same as the one used for the neutron oscillation search.) To our knowledge this is the first measurement of such a spectrum for grafoil material.

The position of the peak at 550 m s^{-1} in fig. 2 is determined by the large spacing of the basal planes of grafoil, with a weak second harmonic visible at twice the velocity. The bulk of the scattering function lies beyond the ordinary graphite Bragg cutoff velocity in the upper part of the cold neutron velocity spectrum. A large velocity-independent component of the cross section, as it would be expected when a strong incoherent scatterer like hydrogen was present, is not visible in fig. 2, nor is there much room for a $1/v$ component from a strongly absorbing impurity.

The absolute size of the scattering cross section shown in fig. 2 was again much higher than the precisely known limiting value of 4.7 b, but it turned out

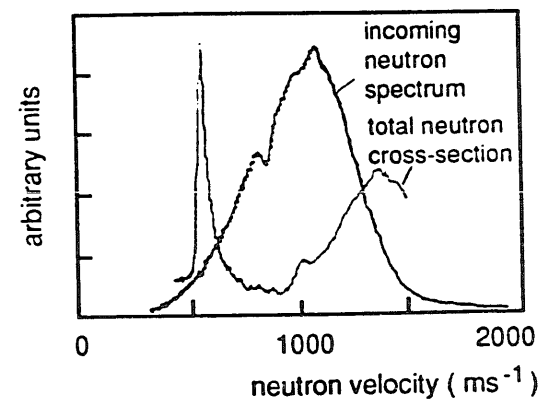

Fig. 2. Measured neutron velocity dependence of the neutron total cross section of exfoliated graphite used as target material. The incoming neutron velocity spectrum is also shown.

that this was an artifact of our transmission method: electron microscopy of grafoil revealed that the material has sufficient structure on a mesoscopic scale on which the neutrons diffract; the resulting small angle scattering then leads to additional neutron losses in the rather distant neutron detector. So we found no reasons to doubt the validity of our prediction (2) on the scattered neutron flux.

The tightness of the large ^{6}LiF shield against scattered neutrons was tested in total neutron transmission experiments done with the ^{6}LiF tiles. For a usual cold neutron beam the transmission was measured to be $\langle T \rangle = 4 \times 10^{-4}$, which is an order of magnitude larger than anticipated in (3). This is due to the inevitable 20% thickness variations within each tile, which enter in the exponent of T. Furthermore, fig. 2 shows us that the (elastically) scattered neutron flux seen by the ^{6}LiF shield has a much "harder" spectrum than the initial flux. When this effect is taken into account then the expected transmission goed further up to about $\langle T \rangle = 2 \times 10^{-3}$, which changes the scattered neutron intensity transmitted through the ^{6}LiF shield, eq. (3), to

$$\Phi_n^{\text{trans}}(^6\text{LiF}) = 1.5 \times 10^6 \text{ s}^{-1} \quad \text{(improved calc.)}. \quad (6)$$

This neutron leakage was also directly measured with a ^{3}He neutron detector which could be moved all along through the nn̄ detector just outside the neutron beam tube. From this measurement we obtained

$$\Phi_n^{\text{trans}}(^6\text{LiF}) = 2.5 \times 10^6 \text{ s}^{-1} \quad \text{(meas.)}, \quad (7)$$

in reasonable agreement with (6). From neutron diffusion calculations we obtain that about 8% of the neutrons should be absorbed by the 2 g cm^{-2} thick trigger plastic scintillators of the nn̄ detector, mostly in hydrogen.

When an additional 2 mm layer of B_4C was added the neutron leakage intensity dropped to

$$\Phi_n^{\text{trans}}(^6\text{LiF} + B_4C) \sim 3 \times 10^4 \text{ s}^{-1} \quad \text{(meas.)}. \quad (8)$$

This, however, did not change measurably the radiation background level in the nn̄ detector. As the additional background from the B_4C capture gammas is negligible, we conclude that neutron leakage of the ^{6}LiF shield was not a main background source.

The fast neutron intensity originating in the ^{6}LiF shield was measured inside the nn̄ detector with the same ^{3}He detector but surrounded by a moderator of 7.5 cm of paraffin covered with a slow neutron absorber of 0.5 cm of B_4C. The detector was calibrated with the known fast neutron flux from the ^{6}LiF beamstop. The result was

$$\Phi_{\text{fast n}}(^6\text{LiF}) = (3\text{–}6) \times 10^4 \text{ s}^{-1} \quad \text{(meas.)}, \quad (9)$$

in good agreement with (5).

Also, water adsorption in the ^{6}LiF shield is not a problem. Earlier investigations [4] with prompt gamma activation analysis showed that ^{6}LiF tiles freshly sintered had the same low hydrogen gamma intensity as one-year-old tiles and as tiles purposely kept in a humid atmosphere.

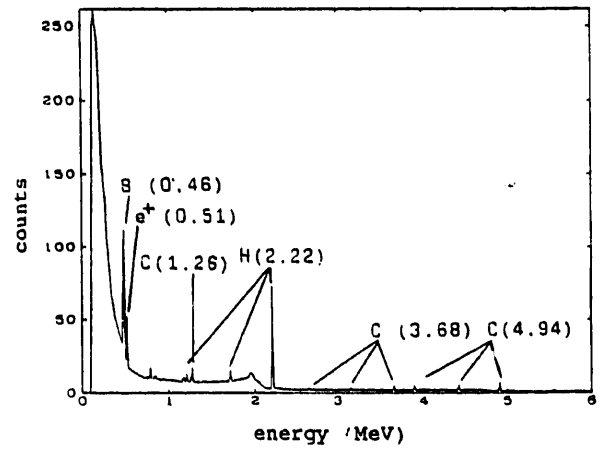

Fig. 3. Prompt neutron capture gamma spectrum of the nn̄ target measured with a Ge (HP) detector. The primary gamma ray energies are given in parentheses (in MeV). The boron peak comes from the B_4C neutron shield, see fig. 1. No target impurities other than hydrogen contribute visibly to the spectrum.

From all these investigations we conclude that neutron scattering from grafoil is not the reason for the enhanced beam-related radiation background. Therefore, we further had to investigate the purity of the target foil, and also of alternative carbon materials. To this end we measured the prompt gamma ray spectra from neutron capture in the target foil with the help of a pure germanium gamma ray detector. This measurement was done twice, first at the ILL's in-beam prompt neutron activation analysis station, and later on directly in the nn̄ experiment with the target in its final position. The prompt neutron activation gamma analysis technique had to be pushed to its limits because pure carbon is already one of the cleanest reactor materials available.

Fig. 3 shows the prompt gamma spectrum of the grafoil target measured with the Ge(HP) detector which was moved through the nn̄ detector in the same way as the ^{3}He neutron detectors. From this spectrum a hydrogen to carbon atomic ratio of about 5% is obtained, which was not included in the impurity list of the supplier, probably because conventional spectroscopy methods are not sensitive to hydrogen impurities.

The hydrogen gamma intensity dropped by a factor of 25 when the Ge(HP) detector was moved from the center of the nn̄ detector to its edge in accordance with the $1/r^2$ law, which proved that the 2.22 MeV hydrogen gammas came from the target region.

Table 1
Radiation background rates induced by neutron–target interactions

Type	Source	Flux [s^{-1}]	See eqs.	Expected backgd. in scintill. [s^{-1}] (at efficiency ϵ)
γ's	target:			
	C	2.2×10^{6}	(1)	1.3×10^{5} ($\epsilon = 6\%$)
	H	$3\text{–}8\times10^{6}$	(10),(1)	$2\text{–}5\times10^{5}$ ($\epsilon = 6\%$)
	other	2×10^{4}		–
	n-shield:			
	^{6}LiF	3×10^{4}	(4)	–
	B_4C	2×10^{6}	(6), (7)	1×10^{4} ($\epsilon = 0.5\%$)
Thermal neutrons	target	3×10^{4}	(8)	–
Fast neutrons	n-shield	5×10^{4}	(5), (9)	–

From our measurements we deduce that the relative hydrogen gamma intensity from the target is

$$\Phi_\gamma(H)/\Phi_\gamma(C) = (2.6 \pm 1.2) \quad \text{(meas.)}. \qquad (10)$$

From the gamma measurements inside the nn̄ detector we also obtained a rough estimate for the absolute value for the total gamma intensity from the carbon target:

$$\Phi_\gamma(C) \approx 6\times10^{6}\ s^{-1} \quad \text{(meas.)}, \qquad (11)$$

which agrees with the more reliable value (1) within a factor of 3.

The hydrogen impurities of about 5 at.% lead to an additional scattered neutron intensity of

$$\Phi_n^{scatt}(H) = (5 \pm 2)\times10^{8}\ s^{-1}, \qquad (12)$$

which is of similar strength to that of the neutron scattering on carbon (2). From the measurement in fig. 2 we had already concluded that hydrogen scattering cannot be dominant. To be compatible with fig. 2 the lower values in the error ranges of (12) and of (10) should be favoured.

Table 1 summarizes our findings. The expected radiation background (single) rates in the inner 2 cm thick layer of plastic scintillators of the nn̄ detector are also listed, at the quoted estimated detection efficiencies. This calculated total noise rate of $(5 \pm 2)\times10^{5}$ s^{-1} agrees with the measured single noise rate of 0.9 MHz, only when the upper values of (10) are favoured. So some inconsistencies remain, although the main issues have been clarified.

We made some attempts to eliminate the hydrogen impurities in the target. As hydrocarbon impurities should be cracked between 2700 and 2800°C we tempered some target material in a high temperature vacuum furnace above 3000°C. After this the hydrogen intensity at 2.22 MeV, measured at the neutron activation station, decreased measurably. The material, however, became very brittle and punctured and no longer fulfilled the requirement of mechanical stability. A high purity carbon monochromator crystal was also tested. The relative intensity of its hydrogen gamma line was only about one sixth that of grafoil. It would, however, be difficult to fabricate a large thin target from this material. A number of other carbon products were also tested, like carbon wool and carbon tissue, but their hydrogen content was even higher than that of grafoil. So we had to use the grafoil target as it was. Finally, an electronic device was developed able to count the total number of tubes [6]. In this was the trigger rate in the experiment resulted in 4 Hz, comparable with the beam off rate.

4. The fabrication of the ^{6}LiF shield

For the large ^{6}LiF neutron shield shown in fig. 1 we first had to transform metallic ^{6}Li into a powder of ^{6}LiF and then produce large thin tiles of sintered ^{6}LiF.

To obtain the ^{6}LiF powder we started from 20 kg of 96%-enriched metal, which was kindly lent to us by the Oak Ridge National Laboratory. Ten gram pieces were dissolved, each in a beaker of distilled water, to give lithium lye. The beakers had to be water-cooled because of the violent exothermal reaction, and care was taken to ventilate away the important amount of hydrogen escaping from the solution. The lye was then neutralized with hydrofluoric acid, leading to the precipitation of fine ^{6}LiF powder. The precipitate was filtered and the powder dried at 200°C and ground. Altogether 80 kg of ^{6}LiF powder were produced in this way.

For the production of our very thin, large and even ^{6}Li tiles the usual method of prepressing the powder and subsequent tempering was not applicable. Instead the dry powder was pressed into tiles in a mould and baked under pressure. The demountable moulds were made of brass, with a square inner section, holding 10 to 20 tiles depending on the desired thickness. For each tile 60 g of powder were evenly distributed in the

mould. Each layer was separated from the walls of the mould and from the previous layer by heat-resistant paper. The layers were compressed with a force of 50 kN in a hydraulic press. The press acted on the cover of the mould which then was bolted tightly in order to maintain the applied pressure.

The moulds were heated in a 3 kW ceramics furnace to 720°C in air for one hour and allowed to cool for 16 hours. Then the sintered tiles were taken out and separated from the heat-resistant paper. Two types of tiles were fabricated, both weighing 60 g: type-I, about 98×98 mm^2 wide and on average 3 mm thick, for the beam dump, and type-II, about 148×148 mm^2 wide and on average 1.3 mm thick, for the protective shield inside the detector. Altogether about 900 tiles were obtained this way. Broken tiles were ground to fine powder, vacuum-dried and recycled. The density of the sintered ^{6}LiF in the tiles is about 85% of the single crystal density.

In the target zone (fig. 1) the wall of the vacuum tube is covered on the vacuum side with the thinner type-II tiles. To economize on the rare ^{6}LiF material the tiles more distant from the target are inclined under an angle of up to 65°.

The beam dump consists of the 140 cm diameter stainless steel vacuum flange onto which the type-I tiles are glued with silicon rubber glue, in such a way that no holes are apparent when looked at from the target. In order to achieve this the tiles had to be placed with an overlap of about 10%. A subsequent 10 cm layer of paraffin and 4 mm of B_4C protects the surroundings from the fast neutrons produced in the ^{6}LiF.

The ^{6}LiF tiles of the beam dump, which are initially white, change their colour under neutron radiation. They become yellow after integration of about 10^{13} n/cm^2, and dark brown after further irradiation. This colouration, which is due to the formation of point defects (colour centers) in the ^{6}LiF crystallites, serves as an additional check on the correct size and position of the cold neutron beam.

5. Conclusion

We showed that the radiation background near a very strong and wide cold neutron beam can be suppressed to a level of 10^{-5}. In the neutron–antineutron oscillation experiment the main residual beam-related background was due to the thin annihilation target foil made of exfoliated graphite and came from an irreducible hydrogen impurity in the carbon target with a 5% atomic ratio. This background was about three times stronger than the inevitable background from the carbon capture gamma rays. The neutrons scattered off the target could effectively be suppressed by a large shield of very thin sintered ^{6}LiF ceramics.

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