Neutrons in contemporary science

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Even before its discovery, the neutron was awaited as a kind of "necessity" in the context of rapidly evolving nuclear physics. In an extraordinary paper cited by Chadwick in his Nobel lecture of dec.1935, Ernest Rutherford predicts the probable existence of a neutral "compound" as the result of fusion of a proton and an electron. Following this assumption, he predicts almost all the properties of the future "neutron":

- a) difficulty to be detected;
- b) zero electric field "except at very close distances";
- c) high penetrating power of all kind of materials.

Much more, in the same paper Rutherford foresees the central role this neutral particle should play in the structure of the nucleus, adding this incredible sentence, that the neutron is probably the true explanation of the stability of all nuclei (!)

All of these predictions prove to be true after Chadwick's pioneering work of 1932. He was the first to "see" secondary tracks of protons and nitrogen ions in a Wilson chamber, leading to the calculation of the neutron's mass, evaluated at about 0,9 times the mass of the proton. In the first part of the lecture, we will go through theses exciting years of discoveries, until the work of experimentalists finally able to see even inside the neutron, again through scattering experiments, leading rapidly to the parton model of the nucleus, soon followed by the quark model of Gell-Mann.

At the end of Part A, we present the most important nuclear reactions involved in nearly all systems for neutron detection, activation, elastic reactions on the proton (or heavier) and the most useful inelastic ones, on ${}^{3}He$ and ${}^{10}B$. A detailed application is presented for measurement of cross sections. with an unknown neutron beam and after calibration of a fast on-line detection system.

Part B of the lecture is devoted to much more recents advances in neutron physics, with highlights on the neutrino mass, on the production of cold neutrons and application of these to exciting items on fundamental thema like electric dipole moment or parity violation.

Finally, I'll present a quite confidential therapy for cancer based on neutron beams (BCNT). Saftey concerns at these facilities make the link to radioprotection general and more generally to neutron dosimetry, illustrated by one of our intrumental developments at IPHC Strasbourg, the AlphaRad chip. Together with our innovative Recoil Proton Telescope, a metrologic tool also based on CMOS pixel sensors, I will conclude by presenting a complete project for a medical application in therapy rooms (radio- as well as hadrontherapy).

Part A Everything is History

1 Early days: before Chadwick

1.1 Rutherford, the pioneer

Ernest Rutherford became a world celebrity through his famous experiment of 1909, revealing the small size of the nucleus. Surprisingly, Rutherford played also an eminent role in the discovery of the neutron. His masterpiece of experimental work was performed only with natural sources, but his scattering setup became a kind of paradigm for all future nuclear scientists. In 1919, still without any accelerating device, he realizes the first transmutation in history through

$$\alpha + {}^{14}N \to p + X$$

The proton was identified thanks to its long range (much higher than the maximal range of an α) and therefore, the unknown X species was, beyond any reasonable doubt, an oxygen nucleus. This achievement was not lead transmuted into gold, of course, but for sure a giant step for nuclear physics. In his paper, Rutherford points the difficulty for charged particles to penetrate the Coulomb barrier without any accelerating device, and he calls litterally for the existence of a neutral particle for which the penetration of charged nuclei would be much easier. Imaginating this hypothetic particle, he predicts the long list of all its properties and he calls really for its existence, in order to explain the stability of all nuclei. Since then, we know how much this is true, starting from the deuteron ${}^{2}D$ until the heaviest ones $(N \gg Z)$ when stability requires more and more neutrons to overcome electrostatic repulsion at the end of the chart.

The complete text of this remarkable prediction can be found in Chadwicks Nobel lecture [1]. In a kind of miraculous inspiration, Rutherford mentions that these neutral particles would be impossible to be contained in a vessel. Even this prediction was found to be largely correct...until 1959, when the extraordinary properties of ultra-cold neutrons were discovered (see the section UCN in Part B).

1.2 Joliot: a historical mistake

Besides Cambridge in the 1930's, the "Institut du Radium" in Paris was another research center for nuclear physics, since the pioneering work of H.Becquerel, Pierre and M.Curie and later, Frederic Joliot, who married Irene, daughter of Marie Curie. Among others, F.Joliot was faced to the mystery of the so-called "beryl radiation", obtained when a strong alpha source is sealed in beryllium walls. The emerging radiation is extremely penetrant, being able to eject protons out of a paraffin screen.

And here comes the first big mistake in this story: Joliot believes that this very penetrating unknown radiation is "probably" made of some highly energetic gamma photons. This opinion was mocked as completely ridiculous by E.Majorana, and in fact, writing down momentum and energy conservation, is is quite simple to check that a photon hitting a proton and able to transfer some 5 MeV to him would have an energy of...100 MeV.

Having neglected to do this simple calculation, Joliot misses the discovery. But the french physicist goes on and on, bombarding everything with alpha particles. At the end, he reaches a completely unexpected result, the β^+ emission of aluminium.

2 Chadwick, then Heisenberg and Yukawa

Obsessed by the real nature of the "beryl radiation", Chadwick follows his own idea, to put the paraffin screen <u>inside</u> a Wilson chamber in order to see something. Doing this, he was able to record the recoils of protons and also of nitrogen ions, these ones firmly identified through the right angles between incoming ion and scattered one, as it has to be for particles of the same mass. Chadwick ignores both the mass and the velocity of the X particles, but a nice reasoning on the maximal observed ranges leads to useful relations between maximal recoil energies and masses. At the end, the ratio observed between nitrogen and hydrogen recoil velocities leads him to this remarkable conclusion: the neutral X particle's mass is 0, 9 time the mass of the proton.

Chadwick ends his Nobel conference by anticipating that the nuclear force is probably of the exchange type. In the same year 1932, W.Heisenberg takes immediately advantage of the discovery of the neutron: his very first nuclear model invoques a unique particle, the "nucleon", which exists in two different states of a new quantum number, the isospin. In this scheme, the proton is the (1,0) and the neutron the (0,1) state of the same particle, the quantum exchange resulting in a stable bound state.

At the same moment but quite far away from Europe, Ideki Yukawa works on bulding a detailed gauge model, based on the exchange of a triplet of bosons of mass 140 MeV. But at these times Japan is isolated from the rest of the world, and Yukawa's work was recognized only in 1949 (Nobel prize). However, these pioneering ideas will prove to be completely right in the 1950's: as powerful accelerators are now available, detailed (n, p) scattering up to 400 MeV [2] reveal indirectly the existence of the pions, several years before their discovery as free particles in cosmic rays.

3 Fermi, Hahn, Meitner and...Oppenheimer

During 1934-1935, E.Fermi follows Rutherford's advice, that neutrons should penetrate much more very easily all kind of nuclei, and in fact they do! Fermi discovers that under neutrons, all the nuclei of the chart undergo β^+ desintegration. In the next years, other surprises will emerge from this intensive work, as for instance al the transuranian species Np, Pu...

In september 12th of 1933, Rutherford gave an interview to the NY Herald Tribune (slide 19, part A), in which he denied the possibility of extracting the huge energy contained in the small nucleus, an idea he calls "absurd". In this paper, his inspiration was not of the same quality than earlier, but the mistake is easy to understand, since fission has not yet been discovered, with the extraordinary feature of additionnal neutrons created on each fission, these ones able to trigger in a fraction of second a complete chain reaction. In fact the discovery of fission was a chemist's game, and Otto Hahn didn't understand at all how baryum could appear in his uranyle salts. In january of 1939, many german scientists have already left their country, and

Lise Meitner is in exile in Sweden. After a letter of Hahn (he speaks of "non-sense") and a visit of Otto Frisch in Göteborg, Meitner understands that the first fission of a nucleus has been observed. Unfortunately, world war II brakes out a couple of months later and all the publications on the subject are classified as top secret. Huge and fast progress are made during the Manhattan Project, but all these results will be published after 1945.

4 A prince among physicists

A couple of years earlier, the big thing in physics was the invention of Quantum Mechanics, which, in his earliest stage, was a simple wave mechanics (1923, De Broglie). Starting from the photon and trying to unify the way physicists describe waves and particles, the frenchman (and prince !) Louis De Broglie postulated a simple relationship between wavelength and momentum of all kind of massive particles:

$$\lambda = h/p \tag{1}$$

This equation was readily verified for electrons and this lead to extraordinary developments, both for theory (Schrödinger Equation) and for instrumentation (electronic microscope). Putting now the mass of the neutron in it, this quite simple equation is already illuminating. For instance, a neutron of 8 *MeV* has a wavelength of 10 fm (10⁻¹⁴ m) which is exactly the scale of the nucleus. An in effect, in diffusion experiments of such fast neutrons on all kinds of nuclei, one gets a wonderful linear relation between the squareroot of the cross section and the power 1/3 of the mass number A of the target. Assuming a hard spheres collision, we can write $\sigma = \pi \cdot r^2$, and this is an elegant way to demonstrate the famous $r = r_0 \cdot A^{1/3}$ (slide 25).

4.1 Condensed matter physics

Taking as another neutron wavelength $\lambda = 1 nm$, De Broglie tells us that the associated energy is about 0.8 meV (yes, milli-eV). And so, in order to obtain diffraction patterns on crystals for example, we need "slow" neutrons.

Diffraction peaks are obtained in the following way: as total spin is conserved, we have the general case of non-zero spin, here no constraint and the diffusion *probabilities* add up uncoherently. But we can also polarize the incoming neutrons, and scatter on magnetic targets (like cubic MnO, anti-ferromagnetic). In this case, it is possible to prepare the system in the initial state $J_{tot} = \vec{S_n} + \vec{S_{Mn}} = \vec{0}$. This time, the outgoing neutron must have spin up (or spin down with one single Mn flipped). In this cas, the coherent sum of amplitudes leads to nice peaks (above uncoherent background) and these peaks can be studied as a function of temperature for instance (slide 28).

Being a De Broglie object, the neutronic wave can also give rise to interference experiments: a classical set-up is the Mach-Zender interferometer, where the incoming beam is split into two secondary beams with appropriate splitting crystals. After recombination, the detecting system is simply translated or rotated and the counting rate exhibits nice interference patterns. Such experiments are used for instance to measure the behaviour of neutrons in the local gravity potential of the Earth [3].

4.2 Thermal spectrum

Unlike protons or other charged particles, neutrons cannot be accelerated and therefore, they are not naturally available in the form of monoenergetic beams. Most of the intense sources of neutrons arise from reactors, where they undergo a thermalization with the water molecules (water is used as moderator as well as for heat transport). Here we introduce the well-known relationship between temperature and mean velocity given by the Maxwell-Boltzmann statistics. The tradition for neutrons is to consider the most probable value, not the RMS, hence:

$$v - n = \sqrt{2kT/m} \tag{2}$$

Putting again some numbers into the game, a source temperature of $T = 300 \ K$ converts into a neutron velocity of 2.2 km/s or 26 meV. For obvious reasons now, the meV - eV region is is the so-called thermal region of neutron energy spectra. These neutrons are not well suited for diffraction on molecular or cristalline structures: as can be verified using eq.(1) and (2), one needs mK neutrons to diffract on 40 nm structures.

Going to even lower temperatures would lead us to "ultra-cold" neutrons, but this is a very special subject and we'll come back to it later on in Part B of this course.

5 Slowing down: why and how ?

Slowing down of neutrons is widely used in nuclear reactors, were the extra-emitted neutrons in each fission process have typically 2 MeV of kinetic energy, and the cross section for further fissions is enhanced by a factor in excess of hundred if we go from the MeV region to "epithermal" of even "thermal" region. The slowing down of neutrons is possible thanks to the simple laws of physics governing elastic collisions. Always conserved is the momentum, if in addition the collison is elastic, then we have kinetic energy as a new conserved parameter. For this kind of collisons and for equal masses $(m_p = m_n)$, the combined relations $\vec{v_1} = \vec{V_1} + \vec{V_2}$ and $v_1^2 = V_1^2 + V_2^2$ lead to the important result that the angle between outgoing proton and neutron (non-relativistic regime) is exactly $\theta_p + \theta_n = 90^\circ$. From this, it follows that:

$$E_p = E_n \cdot \cos^2 \theta_p \tag{3}$$

As the mean value of a cosine square is 1/2, we see that the mean energy of the recoil proton is $E_n/2$, or, in other words, that for each collision, E_n is divided by a factor 2 (mean value).

Another remarkable feature of the billiard game between neutrons and protons is the shape of the recoil distribution. Considering a hard spheres collision, one sees, at the contact between spheres of radius R, that in the LAB reference frame $sin(\theta_p) = b/2R$ where b is the impact parameter. A uniform incoming neutron distribution is obtained if $f(b^2)db^2$ is uniform, therefore $sin^2(\theta_p)$ must be uniform (be careful, neither θ_p nor $sin(\theta_p)$ are uniform !). As a consequence, $cos^2(\theta_p)$ is uniformly distributed, and, according to (3), the distribution of E_p is also flat. Of course, all this is equivalent to suppose isotropy if the center-of-mass frame.

Going to deeper details, we can write the recoil energy of a nucleus of mass A as $E_A = E_n \cdot \cos^2(\theta) \cdot \eta$ with $\eta = 4m_n m_A/(m_n + m_A)$, which is equal to 1 for protons or, on the contrary, is much lower than 1 for more massive target nuclei. After the first collision, the remaining neutron energy E'_n/E_n is in the range $[(A-1)^2/(A+1)^2; 1]$. This mass ratio appears

once again when introducing the *lethargy change*, defined as $u(\theta) = Ln(E_n/E'_n)$. One demonstrates that the mean value $\langle u \rangle$ equals $1 + (A - 1)^2/2A \cdot Ln(A - 1)/(A + 1) = \xi$ which is a constant! As an example for carbon: A = 12, we calculate $\xi = 0.158$, hence the number of collisions $N_{coll} = u/\xi = 111$ needed to lower the neutron initial energy 1 MeV down to 1/40 eV (thermal region).

6 The wonderful deuteron

Having considered diffusion states with neutrons as beams, we can go now to bound states, and first of all to the simplest bound state of nuclear physics, the (n, p) system. The interesting points here are 1) that the deuteron has a single bound state at $E = -2.2 \ MeV$, and 2) that E_1 and E_2 have to be considered together for the proper calculation of the total diffusion cross section. The existence of a single level of bound state can be calculated by elementary Quantum Mechanics with a simple (half-)square potential. Symmetry considerations imply that the wave function must be an odd function, such as to be zero at r = 0 (the reduced particle moves in a half square potential going to infinite for r = 0, which means zero distance between the two nucleons in the true symmetric potential). An experimental check of this is given by the photodissociation reaction $\gamma + n \rightarrow n + p$, which is the inverse of the fusion reaction. The threshold of the photodissociation is at 2.225 MeV and the cross section follows nicely the theoretical calculation (slide 31).

However, even with the correct values of the square potential $(D = -35 \ MeV, w = 2 \ fm)$ the total (n, p) diffusion remained a kind of puzzle: the calculated $\sigma = 3, 5 \ b$ whereas $\sigma = 20.36 \ b$ as measured at $E_n = 1 \ keV$. The diffusion is also readily calculated and such a large discrepancy would be a big failure of nuclear physics. One starts with a pure radial wave

$$\Psi = u(r)/r = e^{ikx} + f(\theta)e^{ikr}/r$$

At l = 0, we can introduce the phase shift δ and so:

$$f_0 = [e^{2i\delta_0} - 1]/2ik = e^{i\delta_0}.sin(\delta_0)/k$$

and we find the cross section

$$\sigma = 4\pi |f_0|^2 = 4\pi \sin^2(\delta_0)/k^2$$

At the low energy limit, we must have $\delta_0 \to 0$ and $f_0 \to \delta_0/k$ which is denoted -a, the "scattering length" [4]. To be noticed, "a" may be positive (bound state) or negative (unbound), and σ is always $4\pi a^2$, calculated to be 3.5 b at low energy. At this point, Wigner asked [5][6]: "Why should singlet ant triplet (bound) state be of the same amplitude?" In effect, with unpolarized neutrons, $\sigma_0 = 3/4({}^3\sigma_0) + 1/4({}^1\sigma_0)$ as 2S + 1 = 3 if S = 1, so we must suppose ${}^1\sigma_0 \gg {}^3\sigma_0$. In other words, the interaction is strongly spin dependant (NB: the bound state is the one with parallel spins, hence anti-parallel magnetic moments, see this paragraph). In his paper of 1940, Wigner proposed a test on ortho- and parahydrogen with epithermal neutrons, and it appeared clearly that: (i)the singlet state is unbound (${}^1a < 0$), (ii) the nuclear forces are strongly spin-dependant and finally, (iii) that the spin of the neutron is exactly 1/2.

7 What exactly is nuclear force ?

At the end of his Nobel Conference, Chadwick announces in 1935 that too few is known about these forces, that they might be of the exchange type and that he and his colleague Feather are going to study collisions of neutrons and protons to learn more about it. In 1951 accelerators are finally available, and Segre et al. demonstrate the existence of exchange forces, becoming visible at about 130 MeV (slide 35). The differential angular cross section shows clearly two contributions, one at low CM angle (attributed to the neutral π^0) and the other one at high CM angles (up to 180°), revealing the transformation of neutrons into protons and vice versa. This additional channel is mediated by charged pions, all these three bosons having a mass of about 140 MeV.

In 1932, soon after Chadwick's discovery, Heisenberg proposed his isospin model, a purely formal symmetry "exchanging" a new quantum number between neutrons and protons and generating stability. Isospin had a kind of success, as it predicted for instance the existence of mirror nuclei. But the exchange model of Yukawa is much more precise, a boson (triplet) is exchanged, and its mass is known. The road to gauge theories is now wide open, and the first free pions are detected in cosmic jets by Powell in 1957.

8 Inside the neutron !

In his luminous paper of 1920, Rutherford stressed that the "unknown neutron" would have vanishing electric field "except at very close distances". This incredible prediction was finally tested in 1961 by Kendall, Taylor and Friedmann who discovered "partons" inside the nucleus. The main achievement of these experiments at SLAC was a precise measurement of the charge distribution density at all distances: the charge density inside protons peaks at some 0.2 fm and goes down slowly to zero, whereas the density inside neutrons peaks also sharply, then it goes negative before reaching zero (slide 37). In fine, this is caused by the value quark content of the nucleons, (u, u, d) and (u, d, d) respectively. Of course, the charge integral is found to be zero for the neutron, this implies a negative part in this distribution. Looking closer to it, the shape if this distribution explains perfectly why the magnetic moments of the neutron is found to be negative! By definition, the magnetic moment is the (integrated) moment of the current density:

$$\vec{\mu} = \int \vec{r} \wedge \vec{j} \ d^3r$$

For the neutron, the negative part of the charge density happens at higher radius, hence a much bigger effect and the net result for the magnetic moment is negative.

Measured values are $\mu_p = +2.79 \ \mu_N$ and $\mu_n = -1.91 \ \mu_N$. Here the unit $\mu_N = e\hbar/2m_p$ is the nuclear magneton. Numerically, $\mu_N = 5 \cdot 10^{-27} \ J/T$, a value 2000 time weaker than the Bohr magneton (mass effect), but il happens that this nuclear magneton is remarkably well suited to NMR experiments with radio wavelengths.

The opposite values of magnetic moments explain why the bound state of the deuteron (n, p) is the triplet one, i.e. parallel spins: this implies anti-parallel moments, as expected.

9 Let's measure cross sections !

What we do today with neutrons is both for theoretical purposes (nuclear models, neutrino physics, ultracold neutrons,...) or for applied science. In any case, one has to calibrate neutron sources as well as neutron detectors: this can be done in two steps as discussed in the following, before going into applications.

The concept of cross section can be derived simply from hard spheres collisions, where the "useful" section seen by an incoming beam is exactly $\sigma = \pi r_N^2$ which goes like $A^{2/3}$. The process is probabilistic: $P = N_{coll}/N_{target} = \sigma/S$ in the thin foil approximation (single collision) and S is the efficient area seen by the beam (beam or target area).

• For a target foil of thickness d, its density is $n_t = N_t/(S \cdot d)$, now we can express the nuclei number much more conveniently as a density: $N_t/S = n_t/d$.

• For a beam of density $\rho(v)$ at velocity v, the fluence is $\Phi_n = \rho(v) \cdot v$ (in $cm^{-2}s^{-1}$). The reaction rate is then the product $R = (\rho \cdot v) \cdot (N_t \cdot \sigma/S) = \Phi_n \cdot n_t \cdot d \cdot \sigma$. One sees that the cross section can be calculated from the measurement of the reaction rate and the knowledge of the beam fluence and the parameters of the target. Practically speaking, one measures rather a counting rate in our γ detector of efficiency times acceptance ϵ , so $C_{\gamma} = R \cdot \epsilon$, and finally:

$$\sigma = C_{\gamma} / (\Phi_n \cdot n_t \cdot d \cdot \epsilon) \tag{4}$$

The complete procedure is first to calibrate the neutron beam, by choosing a well known material for activation (say gold) and an appropriate interval of energy where $\sigma_{activation}$ follows the 1/v-law (see below). If $\sigma_{activation} = K/v$, it follows simply that $R_a = K \cdot \rho \cdot n_t \cdot d$! At this point, the beam is calibrated.

The second step to measure your unknown cross section is to take the new target, select a velocity (chopper, ToF,..), measure the new C and finally deduce the much desired unknown cross section σ_{new} from equation (4).

9.1 The "1/v law"

In 1933, Bohr tried to factorize nuclear cross sections of reactions like A(n, y)B by a model where a compound nucleus is formed before decaying in some final state, leading to $\sigma = \sigma_{comp}\Gamma_y/\Gamma_{tot}$. In 1936, Breit and Wigner proposed their well known formula for resonances of mean energy E_0 and width Γ :

$$\sigma(n,y) = \lambda^2 / 4\pi \cdot 1 / [(E - E_0)^2 + (\Gamma/2)^2] \cdot \Gamma_n \cdot \Gamma_y$$

Fortunately, the Breit-Wigner formula works even far away from resonances! One can evaluate the width Γ_n by a simple density state calculation $\int 4\pi p^2 dp/E$ which goes like v, and with the De Broglie wavelength $\lambda = h/mv$, the result for $\sigma(n, y)$ is a 1/v dependance (side 45).

9.2 A golden target: ^{197}Au

Taking advantage of the very good performances of gamma detectors, activation is one of the most efficient ways to detect neutrons. High precision quantitative measurements are possible with materials like gold or other metals, gold in particular having a full range of very desirable features: (i) the cross section of ${}^{197}Au(n, \gamma)$ follows the 1/v law over decades of energy up to the 4.9 eV resonance; (ii) ${}^{198}Au$ has a very simple decay scheme (Slide 47), a β emission at 295 keV in coïncidence with a dominant γ transition of 411 keV; (iii) a metal easily available in very thin foils (no auto-absorption) and an oxidation-free material. The only drawback of gold, however, cannot be underestimated: its half-life of 2.697 days makes it impossible to calibrate neutron beams directly. For this reason, gold is used as a *primary standard*, and faster detectors can be conveniently calibrated against gold, but out of beam.

A nice example of secondary standard is obtained when thin layers of ${}^{10}B$ are deposited inside gaseous chambers. The reaction (n, α) is instantaneous and such chambers are highly efficient. The uniformity of the ${}^{10}B$ deposit is uneasy to measure, but all what is required is a good calibration of the chamber. This is made very conveniently out of beam, with high accuracy, against a gold foil.

10 The most important reactions for neutron detection

Besides activation (with a very very long list of materials !) the most important reactions for neutron detection are the elastic process (n, p) on hydrogen-rich materials (polyethylene, stearine,..) and the two inelastic reactions ${}^{3}He(n, p)$ and (n, α) on ${}^{10}B$ or ${}^{6}Li$. Fission ionisation chambers are widely used with very thin deposits of ${}^{235}U$, specially useful for low fluences of slow neutrons, and relatively insensitive to gamma radiation.

An important remark has to be made for ${}^{3}He$, which is extremely convenient as gas counter inside Bonner spheres for instance, but this material has become a kind of "rare gas" on earth, therefore more and more expensive! As a proof of this scarcity, the government of China plans to import ${}^{3}He$ directly from the moon's surface (for other purposes of course)...

11 Applications in non-nuclear fields

11.1 Fine arts

Among numerous applications even outside physics, we mention the analysis of historical paintings, usually analyzed with a range of radiations (IR,UV and X) in order to reveal underlying layers, not visible by eye. Neutrons are also used in this field, a technique called NAAR (for Neutron Activation Autoradiography). This method is complementary of the previously mentioned, and NAAR is limited by the availability of neutron sources first, but also by the fact that only a small fraction of the atoms of a painting are sensitive to neutron activation (typically 4 of 10¹²). But only neutrons are able to reveal special pigments like azurite (Cu,Mn), "lead white"" through As,Co and Pb, as well as vermilion (Hg) or P, most of them under opaque layers. A recent reference for this technique is [7].

11.2 Neutrinos

In high energy particle physics, a very nice achievement was the discovery of the neutrino by Cowan and Reines, at the Shavannah River reactor in 1953. Nuclear reactors are huge sources of (anti-)neutrinos, and the historical measurement was made with a special scintillator, optimized for the simultaneous detection of a neutron and a positon, through the inverse neutron decay $\overline{\nu} + p \rightarrow n + e^+$. The neutron had to be detected in coïncidence, therefore a thick scintillator (to obtain nearly 100 % efficiency). The scintillating cristal was H-rich (to slow down the neutron) and doped with gadolinium, such as to detect the activation products (4 γ photons).

The neutrino is still a hot item in physics, with the hierarchy problem still unsolved, and the measurement of the masses going on. The family number is definetly fixed at N = 3 and new constraints on the masses arise from cosmology (Planck's latest results).

11.3 Microelectronics

A very specific activation reaction is used in microelectronics for process control. The standards for these industrial processes is usually put at 4 % (transistors sizes/spacings etc..). Now this industry is running very fast towards nanoscale transistors, hence the requirement of dielectric layers of decreasing thickness in order to keep the same performances. In 2015, the required thickness of these dielectric layers is of some atomic layers, and at this point, the usual oxide SiO_2 is no longer adapted. New exotic materials like Pr od Hf oxides can provide such "high-k" dielectrics, with ϵ constants times 20 ou 40 the value of SiO_2 . One of the best candidates is ¹⁸⁰Hf, for three main reasons: (i) its $\epsilon_r = 24$, (ii) the oxide HfO_2 compatible with the silicon lattice, and also (iii) the fact that once activated ¹⁸¹Hf is a γ emitter at 482 keV. After 4 h irradiation, ultra-thin layers of hafnium oxide can be characterized with 0.92 % precision, well below the requirements of the industry (slide 51, Part A).

Part B

Present day science

In physics, present day interests for neutrons go from pure nuclear physics (halo nuclei) to baryogenesis models or neutron stars. At the level of fundamental quantum mechanics, we aim for instance to get new sources of parity violation. In applied science, we need intense neutron sources for nuclear waste transmutation or generation of radioisotopes for nuclear medicine, and in solid state physics, neutron diffraction is an uncomparable tool for magnetic studies. In the near future, neutron imaging will be applied to luggage control in ports and airports, as X rays interacting with the electron cloud of atoms are easily stopped by thin metal whereas neutrons, interacting only with the nucleus, have a much higher penetrating power.

Going to biology or medicine, I will briefly recall radioprotection principles, with some emphasis on a project of our group at IPHC Strasbourg, the AlphaRad chip. A very special item in the field of medicine is the use of neutrons as a therapy (BCNT), used for twenty years now in Japan.

I will conclude this definitely <u>non</u>-exhaustive review with another project of our group for medical physics based on ultrafast pixel sensors.

12 Neutron sources

12.1 Broad spectra

Neutrons are available at three kinds of places, true radioactive sources, nuclear reactors or accelerators. Among radioactive sources, californium ^{252}Cf is a natural neutron emitter with a maxwellian spectrum peaking around 1 *MeV*. Widely used is the mixed source *AmBe*, a combination as old in history as the famous "beryl radiation" in the 1930's. These broad spectrum sources, of activities in the range of $10^6 n/cm^2/s$, are mainly used for detector calibration.

At reactors, the spectrum is also very large, from very cold neutrons (meV) to some MeV, but here the fluences are orders of magnitude larger. The most powerful source of neutrons for research is the ILL facility in Grenoble (Institut Laue Langevin).

At reactors, one can select energies through Bragg diffraction or mechanical systems:

- Bragg diffraction is a highly selective method, the limitation being the crystal lattice constants: eq.(1) and (2) show that this method is well suited only for quite cold neutrons.

- For energies in the eV range, rotating "choppers" [8] work thanks to the high absorption cross section of cadmium, up to $10^3 b$! Sandwiches of Cd and Al layers (Al being nearly transparent), put in rapid rotation, have been extensively used to scan all neutron energies needed to establish the cross section databases.

- Faster neutrons (in the MeV range) can be produced by accelerators in pulsed-mode, with a time-of-flight setup for energy measurement (n-TOF facility at CERN).

12.2 Monoenergetic beams

True monoenergetic neutron beams can be directly obtained at accelerator facilities, thanks to a limited number of nuclear reactions. One has to choose the appropriate incoming particle, (p,d,..) and its energy, the nature of the target, and the angle of diffusion. This method is limited by the accelerator intensities and target thicknesses (absorption, heating, secondary gamma/electrons,...), but it provides very narrow neutron peaks like for instance the 14.8 MeVline in ${}^{3}H(d, n)$. As this reaction is exactly the one in the ITER project, the tokamak will be full of 14.8 MeV neutrons, and a calibrated beam source at this energy is of highest interest for detector developments in the context of fusion.

Examples at the AMANDE facility [9] located in the Cadarache research center: (p, n) reactions on ${}^{45}Sc$ or ${}^{7}Li$ deliver keV neutrons while 1.2, 2.5, 2.8, 5 and 19 MeV neutrons are provided by protons or deutons on ${}^{2}H$ or ${}^{3}H$.

12.3 Other sources

• Going to high intensity accelerators, a rapidly growing field is the use of <u>spallation</u> reactions which provide important neutron fluences of very high energies (up to the GeV). These neutrons are used for all kind of purposes, nuclear fuel recycling (transmutations), as the (controlled) neutron source of Accelerator Driven Systems of the "Rubbiatron" type. As a proof of the growing interest for such installations, the most powerful spallation source in the world (ESS) is to be built at the Lund University, and to be commissioned in 2025.

• Finally we have to consider unwanted neutron sources, these not only present close or inside nuclear reactors. Its is well known that LHC experiments have to face important neutron backgrounds, these ones generated by the impact of hadronic jets inside the calorimeters of CMS or ATLAS. In these large spectrometers, electronics has to be hardened against two sources of heavy irradiation, charged particles and of course neutrons (up to $10^{14} n/cm^2$).

In a completely different field, conventional <u>radiotherapy</u> sources $({}^{60}Co)$ are more and more replaced by electron accelerator systems in hospitals, and these machines generate a noticable background of neutrons inside the treatment rooms, a subject of increasing concern for nuclear medicine (see last item of the lecture).

13 Ultra-Cold Neutrons: how and what for ?

Coming back to Rutherford's prophetic paper of 1920, one of his predictions was that the hypothetic "neutral object" would be very penetrating and difficult to hold in any kind of vessel. In effect, as long as you consider "normal" neutrons (eV - GeV), their penetrating power in matter is considerable, as Rutherford said, and radioprotection against neutrons is still a difficult task. But suddenly in 1959, something completely unexpected happened.

13.1 From barriers to mirrors and vessels

In 1959, the russian physicist Zeldovitch pointed the fact that below a given temperature [10], very cold neutrons would have such a long De Broglie wavelength that normal metals would appear to them as impassable barriers. At velocities of 10 m/s, as the De Broglie particle has a

wavelength of some 100 nm, a material wall of atomic density N is equivalent to a Fermi potential $U = 2\pi\hbar^2 b_{coh} N/m$, where b_{coh} is the penetrating length inside the material (a couple of fm), depending on the perpendicular velocity of the neutron. If U > 0, the barrier is a true obstacle, and this condition is fulfilled if $v < v_{lim} = \sqrt{2mU}$. Solving the Schrödinger equation leads to a transmission coefficient of zero, hence R = 1 (the perfect mirror) ! Against common sense, such neutrons can really be hold inside a material vessel.

13.2 The iron law of statistics

Numerically, the requested temperature for $v < v_{lim}$ is in the mK range. Inside the big steel vessel of boiling water in a nuclear reactor, the neutron density is of about $10^8 n/cm^3$, this means that the wavelength of most neutrons is well below their mean distance $\lambda \ll \bar{d}$: the neutron gas is therefore fully classic, and the Maxwell-Boltzmann statistics holds. For a mean temperature of 300 K, the mean velocity is 2000 m/s, and the fraction of neutrons of velocity below a given limit velocity is calculated to be $\eta = (1/8)[mv_{lim}^2/kT]^2$. This number is really small, typically 10^{-11} , quite a miserable yield...

If you want to catch UCN neutrons, it works as follows: plunge a cold D_2O target inside the reactor, connect it to your trap through nickel or copper guides, and wait. Only those neutrons which are cold enough will be guided inside your tubes (for the faster ones, the mirror effect just doesn't work!) Once you have some neutrons in your trap, you must be able to run your experiment within minutes, because of the 12 min life-time of free neutrons. In fact the precise measurement of this number is of great importance, being related to the V_{ud} element of the Kobayashi-Maskawa matrix. A good precision has been reached by combining all the cold neutrons experiments all around the world: $885.7 \pm 0.8 \ s$

13.3 A spectacular application: parity violation

Being a perfect quantum object, a cold neutron can be used in a great variety of experiments, either to test quatum mechanics or to be studied as an object for itself. A still open question is to decide if the neutron has a non-vanishing electric dipole moment (EDM). Is the answer appears to be yes, then a new source of parity violation would be accessible, completely independent of K^o or B^o -mesons physics.

The net charge of the neutron is zero, but a permanent electric dipolar moment $\vec{d_n} = e \cdot \vec{r}$ would lead to fascinating consequences at the most fundamental level. The only specific axis for the neutron is its spin direction, so we must have $\vec{d_n} = d_n \cdot \vec{s}$. If now we apply the parity operator, $P(\vec{r}) = -\vec{r}$ but $P(\vec{s}) = \vec{s}$ and so, d_n is a pseudoscalar entity. Nothing in the known laws of physics imposes to the neutron to have non-zero EDM, but the Standard Model puts strigent limits to its possible value, to about $10^{-32} \ e.cm$. The present experimental limit is $2.9 \cdot 10^{-26}$, and UCN could help to push this limit down to exciting new physics, beyond the SM.

The experimental setup for these measurements are combinations of (high) electric fields and small magnetic ones. By measuring spin-flip ratios between parallel and anti-parallel states (Ramsey method), the result is directly sensitive to d_n , with a precision proportional to \sqrt{N} the number of flipped neutrons. To test this new source of parity violation, we only have to increase the statistics, what means building bigger sources of UCN.

14 Neutron detectors

14.1 Only one method !

For detection of neutral species there is only one method: generate a reaction leading to charged secondaries (and this includes activation, as the gamma photons will, *in fine*, generate electrons). For neutrons we can choose elastic processes like (n, p) or inelastic ones like (n, α) on lithium/boron, activation which is the (n, γ) reaction and also fission. Targeting now these charged species, the choice is open between passive systems like SSNTD or, if you want some electronic output, between **1**) gaseous chambers, **2**) scintillators or **3**) semi-conducting devices. At this point, the interested reader has only to refer to classical textbooks on detectors for charged particles...

A historical method in nuclear physics was to use emulsions to detect secondary protons: thanks to the flatness of the energy distribution of the recoil protons in the elastic process (n, p), it is quite easy to reconstruct the incoming E_n , with the obvious inconvenient that only a small fraction of the recoils is used, in the region of maximal recoil. This discussion arises also with modern methods like telescopes (next section).

14.2 Specific systems

• Bonner spheres: the best system to measure a complete (unknown) neutron spectrum is a collection of Bonner spheres of calibrated diameters. The outerpart of each is a moderator (polyethylene) such as to slow down all neutrons for optimal detection near the center of the sphere equipped with a ¹⁰*B*-enriched chamber of the Geiger type. Closely associated to simulation, this system allows a nice reconstruction (after deconvolution) of the complete spectrum [11].

• DEMON: Nuclear physics is more and more concerned in detecting the complete final state of nuclear reactions, including neutrons. In this case, 4π systems of high efficiency are required, such as to detect all components of the final state. Thick cylindrical tanks of liquid scintillators $(30 - 50 \ cm)$ have currently 70 - 80% efficiency even for E_n up to several tens of MeV. As an example, the DEMON system [12], fully modular, can be arranged in vertical walls or in 4π configuration, in association with any other detectors (e.g. $E - \Delta E$) for charged secondaries.

Closely associated to scintillating systems is the lang-standing problem of neutron/ γ discrimination. In effect, all the known sources of neutrons are "contaminated" by gamma photons, and unfortunately, these two neutral particles behave more or less the same way inside any kind of detector. Nuclear physics has to take care of this mimetic behaviour, and especially large scintillators must be operated with a discriminating strategy. The most efficient way to distinguish these two species is to analyze the pulse shape from the photomultipliers. Photoelectrons generated by MeV-gammas and recoil protons from neutrons behave not exactly the same way inside the scintillator, the small and the difference between the two types of pulses can be efficiently used to distinguish both species. Today this is done automatically by dedicated electronic signal treatment [13].

• Dosimetric systems: passive dosimetry consists in patient recording of neutron radiation (over days/weeks), in various types of solid state films/plates/rods, called in a generic way SSND (for

Solid State Nuclear Track Detectors). After conversion, the secondary charged particles are catched for instance in polymer foils (CR39), where they induce quite invisible damages, but easily recorded with optical systems after chemical post-processing [14]. Other systems use different physical principles like thermoluminescence (TLDs) or latent images in imaging plates $(BaFBr : Eu^{2+})$. It has to be noticed that simple systems like polymer films have been used even in sophisticated experiments like UCN.

The three families described above work at best with a careful *simulation*. Even if the secondaries are efficiently detected, central questions are always the same, the true efficiency to neutrons and the possible gamma contamination. Mostly used are the MCNP package [15], and open-source codes like FLUKA and Geant IV.

15 A metrologic device: the CMOS-RPT

A strips/pixels telescope is a standard tool for tracking of relativistic particles, and surprinsingly such an arrangement is also a powerful system for measurement of neutron fluences and energies. The most convenient device is a gaseous ionising chamber equipped with an hydrogen-rich converter at the entrance [16]. The simplest choice here is to select proton tracks aligned with the target direction ("zero degree" telescope). Doing this, a zero degree telescope allows to measure E_n with high precision, as can be seen by derivation of eq.(3):

$$[\sigma(E_n)/E_n]^2 = [\sigma(E_p)/E_p]^2 + 4 \cdot \tan^2(\theta_p) \cdot \sigma^2(\theta_p)$$

Note the remarkable apparition of the *absolute* error on the angle ! Of course taking $\theta_p = 0$ is the faster way to cancel this additional term, and doing this one achieves the same precision on E_n than on the protons energy. But here again, selecting only the zero-degree diffusion is a considerable loss of events, as protons are created at all angles in the CM frame. Therefore, an alternative method is to keep all the secondaries to enhance statistics, but this implies a real tracking of the protons, not a sharp selection in their angular distribution. At energies well above 30 MeV, several tracking systems have been operated around the world, but nothing was available at the energies of the AMANDE facility [1; 20 MeV].

Given the expertise at IPHC Strasbourg on CMOS pixel detectors, we developed a true tracking telescope based on very thin silicon pixels (slides 18 - 19 part B). After the H-rich converter, the CMOS-Recoil Proton Telescope is made of three planes of pixels for precision tracking and a final 3 mm thick diode to catch the remaining proton energy. This way, a simultaneous measurement of θ_p and E_p leads to an excellent reconstruction of E_n through eq.(3). As the pixel sensors are distant of only 6 mm, our RPT has an angular acceptance of 41° . Pixels at 50 μ m pitch offer an adequate precision on tracking given the natural limitation of multiple scattering. As each sensitive plane is thinned down to 50 μ m, very slow protons can travel through the device, and the lowest energy we can detect is $E_n = 5 \text{ MeV}$ [17][18].

A fast version of our telescope is under development, in order to keep control on the inelastic background. These events are generated everywhere inside the device, in coïncidence with true protons. The only way to get rid of them is to enhance the readout speed. For this, a dedicated pixel chip with very fast readout time has been developed by our group, the FastPixN chip [19].

16 Human biology

16.1 Neutron dosimetry in short

In a previous section, specific systems for neutrons have been mentioned, SSNTD, TLD or others, unexpensive and therefore well suited for passive dosimetry. For all these systems however, trying to operate them as dosimeters is by no means straigthforward, and special attention must be paid for reproductibility during pre- and post-processing. Owing to the thinness of these films, their efficiency is quite poor $(10^{-3} p/n)$, but the central point is proper calibration. The three important figures of merit of a good dosimeter are linearity, minimal level of sensitivity and high dynamical range, as the level where the film is completely "black" (saturation) must be well above the legal limits (5 mSv/y for ordinary people).

16.1.1 Almost everything in one curve

Most of these systems are first of all counting devices, generating one secondary for one neutron (even at low yield). The game is just to convert a total counting into a professional dosimetric unit.

• The first specific point concerning the neutron is its weighting factor: to convert a massic dose $(1 \ Gy = 1 \ J/kg)$ into a human-adapted dose H (in Sv), one applies weighting factors in the summation $H = \Sigma \ w_i \cdot D_i$ over all radiation types i such bas to take into account the specific danger of each one. Slide 33 (*part B*) shows that neutrons generate 15-20 times more damages than gamma photons to human/animal cells, hence a conventional $w_n = 20$ for neutrons.

• The second specificity for neutrons is the way one converts fluences into doses. The curve of slide 34 (*part B*) gives the standard calculation to go from the fluence $(n/cm^2/s)$ to an effective dose (*Sv*). A sharp increase (a factor 50 !) is seen for the effect of fast neutrons compared to slow ones. As an example taken from neutrons for radiotherapy, slide 32 (*part B*) shows the deposited dose inside a phantom for human body: taking $10^6 n/cm^2$, this converts into $10 \ \mu Sv$ for slow neutrons but this number jumps to 0.5 mSv for the same fluence in case of fast neutrons !

16.1.2 An IPHC project: The AlphaRad chip

Thanks to the expertise of the IPHC in microelectronics and integrated pixel sensors, we developed a dedicated chip for fast detection of MeV-protons and alpha particles, on other words a potential counting system for fast and thermal neutrons. The AlphaRad chip of 0.5 x 0.5 cm is γ -transparent [20], low power consuming, sensitive to the single proton or alpha (i.e. some μSv of neutrons), with a neutron rate higher than $10^8 \ cm^{-1}s^{-1}$. With all these features [21][22], the chip is a good candidate as a powerful electronic device for operational neutron dosimetry, even in medical environments (see last section).

16.2 Neutrons for medicine

16.2.1 Production of radio-isotopes

Nuclear medicine makes use of radio-isotopes both for imaging and for treatment. Specific gamma emitters like ¹⁸F or ⁹⁹Mo are produced in reactors or in cyclotrons, allowing Positon Electron Tomography (PET) for the first one or 140 keV gamma imaging through the daughter

metastable isotope ${}^{99m}Tc$. Research on this subject is still active, as "good" isotopes must have a short life-time in the body and should emit positons or directly gamma photons at the desired energies. As gamma-diagnosis goes increasing (30 million tests par year in the world) a dedicated source for production of the radioisotope ${}^{99m}Tc$ will be completed in 2020 at Cadarache (Jules Horowitz Reactor).

Brachytherapy is considered as a "radical" treatment method, as highly active needles are brought very close to the tumor through the patient's body. Despite its long life-time (74 d), one of the mostly used isotope here is iridium ¹⁹²Ir, obtained by previous neutron bombardment of the stable element ¹⁹¹Ir.

16.2.2 Neutrons as therapy

The high penetrating power of neutrons has the potential of reaching tumors at all depths inside human bodies. The idea of BNCT (Boron Neutron Capture Therapy) is to send specific molecules close to the tumor [25], and then to generate a kind of brachytherapy triggered from outside by neutrons irradiation through (n, α) . The idea seems wonderful as the range of these alpha particles in water is exactly the size of cells. And very smart molecules can be designed today, with a searching head able to target tumor cells and, at the other end, another structure to convey ¹⁰B for treatment, or ¹¹B for imaging.

However, the first challenge of BNCT molecules is to penetrate *inside* tumors, whose size is sometimes of several tens of cm! The second drawback is the need to put patients directly in neutron beams. As mentioned in the section "dosimetry", only slow neutrons must be selected in order to minimize direct irradiation all along the path of the neutrons. Taking the same number as before $(10^6 \ n/cm^2)$ in the phantom), this instantaneous number must be multiplied by the duration of the exposure. Knowing that a typical BNCT treatment is of several minutes per day, we can see that such fluences of parasitic neutrons are certainly not harmless.

16.3 The IPHC project for radiotherapy

Having developed both the AlphaRad chip (fast counting) and a CMOS-pixel Recoil Proton Telescope (spectrometer), we aim to use these two devices directly during radiotherapy treatment. In effect, high energy electron guns for radiotherapy (16-25 MV) have been proved [23][24] to generate a non-negligible level of secondary neutrons by photo-neutron reactions on the tungsten collimators at the end of the machine. This secondary radiation has to be mesasured carefully before efficient shielding. The Bonner multi-sphere system [11] is heavy and difficult to handle, and in such places one needs a flexible method to characterize the neutron spectrum quickly and easily. This can be done with our fast and compact RPT spectrometer [18][19] with help of a dedicated GEANT simulation. Once the spectrum is known, our fast AlphaRad counters can be used as electronic dosimeters in medical rooms, being small and low power consuming [26], with no need of post-processing and no risk of saturation. An additional possibility of these operational devices is automatic recording for individual patients as well as blind data taking for epidemiologic studies of this unwanted (and potentially dangerous) neutron background.

17 References

- [1] J.Chadwick, Nobel Lecture, dec. 1935 www.Nobelprize.org.
- [2] O.Chamberlain, E.Segre, C.Wigand Phys.Rev. 83 (1951)923.
- [3] J.L.Straudenmann et al., Phys.Rev.A 21,1419 (1980)
- [4] E.Fermi and L.Marshall Phys.Rev. 71 (1947)666.
- [5] J.Schwinger and E. Teller Phys. Rev. 52 (1937)286.
- [6] E. Wigner Phys. Rev. 51 (1937)947.
- [7] L.Monico, M.Alfeld et al. Jou.Analyt.Ato.Spectrosc. 30,3 (2015)613.
- [8] J.R.Dunning et al. Phys.Rev. 48 (1935)704.
- [9] V. Gressier et al. Rad. Prot. Dosim. 110 (2004)49.
- [10] Y.Zeldovitch, **JETP** 36, (1959) 1952.
- [11] V.Lacoste et al. Rad.Prot.Dosim. 110 (2004)529.
- [12] Y.Aritoma et al. Nucl.Phys.A 759 (2005) 309.
- [13] F.Fernandez et al. Rad.Prot.Dosim. 70 (1993)87.
- [14] F.Fernandez et al. Rad.Prot.Dosim. 44 (1992)337.
- [15] A.Nachab, D.Husson et al. Radiation Measurements 40 (2005)275.
- [16] B.R.L.Sievert et al. Nuclear Instrum.Meth.A 235 (1985) 542.
- [17] A.Allaoua, D.Husson et al Rad.Meas. vol.44,9-10 (2009)755.
- [18] J. Taforeau, D. Husson et al. J.Instrum. vol7 (2012).
- [19] M.Kachel, D.Husson et al. Rad.Prot.Dosim. 161,1-4 (2014)249.
- [20] M. Vanstalle, D. Husson et al. Nuc. Instrum. Meth. A 662,1 (2012) 45.
- [21] D.Husson et al. Nuc.Instrum.Methods A 569 (2006)845.
- [22] Y.Zhang, D.Husson et al. Microelectronics Journal 43,11 (2012)730.
- [23] J.Farah, et al. Physics in Medicine and Biology. 59(2014)2747.
- [24] X.G. Xu et al. Physics in Medicine and Biology. 53 (2008).
- [25] T.Aihara et al. Int.Jou.Clin.Oncol. (2013).
- [26] Y.Zhang, D.Husson et al. IEEE Trans.Nuclear Sc. 59,4 (2012)1465.

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