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NUCLEAR ORIENTATION ; FROM AN EXOTIC TO A MATURLED METHOD IN NUCLEAR PHYSICS

H. POSTMA

Natuurkundig Laboratorium, Rijksuniversiteit Groningen, Netherlands

Résumé. — L’orientation nucléaire est appliquée en physique nucléaire depuis plus de deux décades. Considérée tout d’abord comme une méthode un peu exotique in spectroscopie nucléaire, elle est devenue une technique utile dans ce domaine et dans quelques autres de la physique nucléaire. Les possibilités et difficultés actuelles seront examinées de façon assez détaillée. Plusieurs exemples d’expériences d’orientation nucléaire seront passés en revue.

Abstract. — Nuclear orientation has been applied in nuclear physics for over two decades. It started as a somewhat exotic demonstration in nuclear spectroscopy, but it has evolved to a useful technique in nuclear spectroscopy and some other fields of nuclear physics. The current possibilities and difficulties will be considered in some detail. A few examples of nuclear orientation experiments will pass in review. Nuclear orientation can also be useful to obtain information on the nuclear hyperfine interaction in solids.

1. Introduction. — Nuclear orientation is the ordering of nuclear spins in space by an external influence; e.g. a magnetic field, \( H \), coupled with the nuclear magnetic dipole moment, \( \mu \). Due to such a field the degeneracy of the \( 2I + 1 \) sublevels, \( I \) being the nuclear spin, is removed. The energies of these sublevels are given by: \( E_m = - (\mu / I) H m \). With an ensemble of nuclear spins in thermal equilibrium the probabilities, \( a_m \), of finding nuclei in sublevels \( I = m \) are proportional to the Boltzmann factors

\[
\exp(-E_m/kT) = \exp(\mu H m/|kT|)
\]

of course \( \sum_m a_m = 1 \). Consequently there are at most \( 2I \) independent parameters to describe the nuclear orientation. Instead of the \( a_m \) it is much more suitable to use parameters defined as linear combinations of the moments \( \sum_m m^r a_m \) with \( r \) integer. The first order nuclear orientation parameter is given by:

\[
f_1 = I^{-1} \sum_m m a_m .
\]

It plays a role in experiments, which are sensitive to nuclear polarization. That is the emitted radiation or reaction must be able to distinguish between «up» and «down» directions of the nuclear spin. The second orientation parameter, which is of interest to us, is the alignment parameter:

\[
f_2 = I^{-2} \left\{ \sum_m m^2 a_m - \frac{1}{2} I(I + 1) \right\} .
\]

It does not distinguish the « up » and « down » directions. Other higher order odd and even orientation parameters may occur, but they are not of sufficient interest for this talk to write down their exact expressions. The orientation parameters are defined in such a way, that they are zero for non-oriented nuclei; that is when all \( a_m \) are equal.

In order to obtain appreciable differences between the \( a_m \) values, and thus useful values of the above-given orientation parameters, it is necessary that \( \mu H /|kT| \) is of the order of unity. Assuming a nuclear dipole moment of the order of \( 1 \) nuclear magneton \( H/|T| \) should be \( 10^7 \) Oe/K or larger. This is difficult to achieve under laboratory conditions.

2. Historical review. — In 1948 both Gorter \cite{Gorter_1948} and Rose \cite{Rose_1949} proposed to use the strong internal magnetic fields (up to about \( 10^7 \) Oe) in paramagnetic ions to achieve nuclear orientation. These fields were discovered in optical and micro-wave spectra; they are generated at the nuclei by unpaired local electrons. With the aid of adiabatic demagnetization of suitable paramagnetic salts it was possible to obtain temperatures of the order of 0.01 K. Thus in principle the conditions for achieving nuclear orientation were fulfilled for at least a small number of isotopes.

The first successful nuclear orientation experiments were performed by Daniels et al. \cite{Daniels_1960} in Oxford using a single crystal of \( \text{CuSO}_4 \cdot \text{Rb}_2\text{SO}_4 \cdot \text{H}_2\text{O} \) in which some of the copper was replaced by \( \text{^{65}Co} \) and by Poppema et al. \cite{Poppema_1964} in Leyden using also \( \text{^{60}Co} \) in a single crystal of \( (\text{NH}_4)_2(\text{SO}_4)_2 \cdot \text{H}_2\text{O} \). The known gamma transitions of \( \text{^{60}Co} \) were used to detect the orientation. The direct nuclear information obtained concerned the nuclear magnetic dipole moment of \( \text{^{60}Co} \). It was clear from the very beginning that interesting nuclear information...
can be obtained with the aid of nuclear orientation. However, nuclear physicists rather liked to consider nuclear orientation as an exotic tool. The reasons were, that at that time only a few isotopes seemed to be suitable candidates and that the experimental conditions were difficult to achieve, expensive and limited to a few low-temperature laboratories. Moreover, nuclear physicists just learned to use γ-γ angular correlation techniques, which enable to obtain similar information about nuclear levels.

Notwithstanding this tremendous draw-back a few physicists were willing to prove the usefulness of nuclear orientation in certain regions of nuclear physics. In this respect I like to mention as an example the study of \(^{56}\text{Co}\) with orientation techniques in Leyden as a joint effort with the university of Groningen. This experiment [5] gave significant new information about properties of levels and transitions of \(^{56}\text{Fe}\). Very interesting sources of information were the linear [6], [7] and circular [8] polarizations of gamma radiation emitted from oriented nuclei. The determination of neutron resonance spins through the interaction of polarized neutrons and polarized nuclei was soon explored by physicists at Oak Ridge [9].

During the decade following the first demonstration of nuclear orientation the number of isotopes, which had been oriented grew slowly but steadily. Much more became known about the magnetic properties of solid state materials at low temperatures including hyperfine interaction. The coupling of nuclear electric quadrupole moments with electric field gradients in solid materials was successfully applied in a number of cases. All these methods are static in nature, that is, the ensembles of nuclei are in thermal equilibrium with their direct surroundings. Besides these static methods dynamic techniques for orienting nuclei were developed. As examples I only mention the polarization of protons by microwave resonance techniques, which is extensively used in medium and high energy physics, and optical pumping of for instance \(^{3}\text{He}\).

The historic experiment of Wu et al. [10] in 1956 concerning the asymmetry of β-emission from polarized \(^{60}\text{Co}\) nuclei, which demonstrated very clearly the non-conservation of parity in weak interactions, brought nuclear orientation under broader attention.

However, still further technical developments were necessary to enlarge the applicability of nuclear orientation to make it of sufficient interest for a larger group of experimentalists. I will mention some of the new developments and improvements, which made nuclear orientation of more general use during the past few years.

In 1960 an important discovery was made by Samoilov et al. [11] and by Kogan et al. [12] that nuclei of diamagnetic atoms incorporated as impurities in a ferromagnetic material like iron experience strong magnetic fields. By cooling magnetized iron foils, containing radioactive nuclei like \(^{186}\text{Re}\), \(^{192}\text{Ir}\) and \(^{198}\text{Au}\), to temperatures below 0.05 K large anisotropies of gamma radiation were found. Fields as large as 1.6 MOe were derived from such measurements. Only small quantities of radioactive material were allowed to diffuse into the iron lattice. This was quite often very difficult or impossible due to chemical reactions or clustering.

The use of impurity atoms in ferromagnetic hosts became considerably more interesting with the introduction of the implantation technique with the aid of a mass separator. This allows to distribute in a rather uniform manner mass-separated isotopes in thin layers of pure iron foils. This method was first applied by De Waard and Drentje [13] for preparing Mössbauer sources. The use of impurity atoms implanted inside iron became an important tool in nuclear orientation experiments during the past few years.

Another very important development for nuclear orientation is the invention of \(^{3}\text{He}-^{4}\text{He}\) dilution refrigerators, which enable continuous cooling to temperatures as low as 0.01 K, although in practical cases the lowest temperature to be reached is somewhat higher (say 0.03 K) due to the heat input at the samples. Also adiabatic demagnetization allows one to obtain such temperatures but there are two main disadvantages, namely a) the cooling is not continuous and b) the requirements for magnetic fields are usually conflicting. The latter means that sample and coolant should be separated by a relatively large distance. With \(^{3}\text{He}-^{4}\text{He}\) dilution refrigerators low temperatures can be obtained with relative ease. It is also possible to change and stabilize the sample temperature with the aid of a small heater.

Now a days strong magnetic fields can be generated with the aid of superconducting solenoids. This is not yet a very essential development for nuclear orientation, but with new discoveries in superconductivity one might hope that still stronger split-coil magnets can be constructed. This will open new possibilities for nuclear orientation.

It would be very interesting to develop techniques to reach still lower temperatures. Two promising possibilities are a) adiabatic compression [14] of liquid \(^{3}\text{He}\) to solid \(^{3}\text{He}\) and b) adiabatic (nuclear) demagnetization [15] of PrCu, or similar alloys. It seems reasonable to expect that in near future nuclear orientation experiments with samples cooled down to about 0.005 K are becoming popular. With such developments nuclear orientation can be obtained for a very large group of isotopes.

It is, however, important to remark at this point that one formidable problem will remain; namely the heat conductivity which decreases strongly with decreasing temperatures. This will prevent the use of still lower temperatures in nuclear orientation. It is clear that only weak radioactive sources can be used if the heat conductivity becomes the limiting factor. Also this prohibits many experiments with particle beams, with the exception of neutron beams.
After this short sketch of the development of static nuclear orientation techniques I like to consider some of the aspects of hyperfine interactions in relation to nuclear orientation. Then a few examples will be given to elucidate the possibilities of nuclear orientation methods in nuclear physics.

3. Hyperfine interaction and nuclear orientation. — Let us consider the simple case of the coupling of a nuclear dipole moment with a magnetic field along a well-defined direction. The interaction Hamiltonian is given by:

\[ \mathcal{H} = - \mu \cdot \mathbf{H} = - g_N \mu_N \mathbf{H} \cdot \mathbf{I}, \]  

in which \( g_N \) is the nuclear gyromagnetic ratio. This gives a set of equidistant sublevels with \( m \) varying from \(-I\) to \(+I\). Introducing \( \beta = \mu H / kT \) the degree of polarization is given by:

\[ f_1 = \frac{2I + 1}{I} \coth \frac{2I + 1}{I} \beta - \frac{1}{2I} \coth \frac{1}{2} \beta \]  

which equals in the high-temperature approximation to:

\[ f_1 = \frac{1}{3} (I + 1) \beta = \frac{1}{3} \frac{I + 1}{I} \frac{\mu H}{kT}. \]  

The alignment parameter is given by:

\[ f_2 = \frac{2}{3} \frac{I + 1}{I} - f_1 \coth \frac{1}{2} \beta \]  

and in the high temperature approximation:

\[ f_2 = \frac{(I + 1)(2I - 1)(2I + 3)}{90 I^3} \left( \frac{\mu H}{kT} \right)^2. \]  

Thus the polarization is in first order proportional to the reciprocal temperature, while \( f_2 \) shows a high-temperature dependence proportional to \( T^{-2} \). Hence at intermediate temperatures \( f_2 \) will be small, while \( f_1 \) has already a rather large value. Thus polarization experiments can often be carried out at somewhat higher temperatures compared to experiments which depend on the alignment parameter.

In a number of cases orientation is achieved by the coupling of nuclear quadrupole moments with electric field gradients, \( \partial^2 V / \partial z^2 \), in crystals. Under the assumption of rotational symmetry the interaction Hamiltonian is given by:

\[ \mathcal{H} = \mathcal{P} \{ I_z^2 - \frac{1}{2} I(I + 1) \}. \]  

where

\[ \mathcal{P} = \frac{3 eQ \partial^2 V / \partial z^2}{4 I(2I - 1)}. \]  

The hyperfine levels are split in a number of doublets, \( \pm m \), which are not equidistant. It is clear that the nuclei do not polarize; it is typically a method for aligning nuclei.

A well-known case for nuclear orientation by electric quadrupole coupling is \( ^{235} \text{U} \) in the uranyl group \( \text{UO}_2 \).

In paramagnetic substances at low temperatures the interaction can be written in terms of an effective spin \( S' \). In case of rotational symmetry and neglecting direct coupling with the external field the interaction Hamiltonian can be written as:

\[ \mathcal{H} = D \{ S_z'^2 - \frac{1}{2} S'(S' + 1) \} + g_\text{n} \mu_B H_z S'_z + \]

\[ + g_\perp \mu_B (H_x S'_x + H_y S'_y) + AS'_z I_z \]

\[ + B(S'_z I_x + S'_y I_y) + P \{ I_z^2 - \frac{3}{2} I(I + 1) \}. \]  

The first term gives the splitting of the electronic multiplet by the electric crystal field if this is not very much larger than the hyperfine splitting. In such cases an effective spin larger than \( 1/2 \) is defined. In many cases an effective spin \( S' = 1/2 \) can be used. The second and the third terms give the Zeeman splitting. The last term is the electric quadrupole coupling term, while the \( A \)- and \( B \)-terms depict the magnetic hyperfine coupling.

Without a magnetic field and with \( B \) and \( P \) negligibly small compared to \( A \), we are dealing with doublets \( (S'_z = 1/2, I_z = m \) and \( S'_z = -1/2, I_z = -m \) which are equidistant. This will lead to alignment according to formula 5 in which \( \beta = A/2kT \), and \( f_1 \) is the absolute value for the polarization in each of the subsets with \( S'_z = 1/2 \) or \(-1/2 \). The \( B \)-term mixes sublevels with \( m \)-values which differ \( \pm 1 \). This reduces the alignment parameter. With \( B > A \) a negative value of \( f_2 \) is obtained; that means the nuclei tend to orient in the \((x, y)\) plane.

If a magnetic field is applied the two-fold degeneracy is removed. The nuclei will tend to polarize along the quantization axis. In addition the influence of the \( B \)-term is reduced.

4. Gamma-ray spectroscopy. — Many of the nuclear orientation experiments concern the emission of gamma radiation, of which the angular distribution is given by:

\[ W(\theta) = 1 + \sum_{k = 2; 4; \ldots} f_k(1) G_k A_k P_k(\cos \theta). \]  

where \( \theta \) is the angle of emission with respect to the orientation axis, \( f_k \) are the even orientation parameters, \( G_k \) are parameters depending on preceding transitions and the \( A_k \) depend on properties of the transition under consideration (initial and final spin, multipole orders and multipole mixing). In most cases only terms with \( k = 2 \) and 4 have to be considered.

If the decay properties are sufficiently well known it is possible to derive \( f_2(1) \), neglecting higher order terms for the time being from the anisotropy measurements as a function of temperature. If the magnetic field is known in the case of magnetic hyperfine interaction, it is possible to obtain the magnetic moment of the decaying nucleus. Such experiments have
been carried out recently in Louvain in order to study the trend of the magnetic moments of \(3/2^+\) and \(11/2^-\) states in the tellurium region [16]. If orientation is due to electric quadrupole coupling it is in principle possible to obtain the nuclear quadrupole moment.

The problem is then that the electric field gradient must be known either from calculations or from other experiments.

In general the anisotropy of \(\gamma\)-emission from oriented nuclei will be used to extract information about spins of intermediate levels and about multipole orders and multipole mixing. Not all of this information can be obtained from a single experiment. Other experiments, such as \(\gamma-\gamma\) measurements or electron conversion data, are necessary for this purpose. As an example I like to refer to a series of experiments with \(^{193}\text{Ir}\) and \(^{194}\text{Ir}\) isotopes in iron with 1\(^{\text{st}}\) iridium and oriented at low temperatures reached by adiabatic demagnetization of a chrome-potassium pill. These experiments were carried out by Reid et al. [17] in order to derive the nuclear moments of \(^{193}\text{Ir}\) and \(^{194}\text{Ir}\) and to study levels populated in the decays of these isotopes.

An interesting case is the study of \(\gamma\)-rays emitted after capture of polarized slow neutrons by polarized nuclei. Then the angular distribution is given by:

\[
W(\theta) = 1 + A_{11}^0 f_1(n) f_1(N) + A_{22}^0 f_2(n) f_2(N) P_2(\cos \theta),
\]

where \(f_1(n)\) is the neutron polarization; \(f_1(N)\) and \(f_2(N)\) are the nuclear polarization and alignment parameters. This expression holds for gamma radiation directly emitted from the capturing state. Such transitions are usually of dipole character, which means, that only orientation parameters up to the second order have to be considered. The two polarization parameters \(f_1(n)\) and \(f_1(N)\) occur as a product. The second term expresses the change in partial capture cross section for a particular \(\gamma\)-transition. Slow neutron capture is an s-wave process, hence the compound state has spin \(J = I - 1/2\) or \(I + 1/2\). It quite often happens, that at thermal energy cross sections related with both compound spins overlap. As a consequence interference effects may occur.

In the above-given equation \(A_{11}^0\) depends linearly on the fraction of \(I - 1/2\) capture, while \(A_{22}^0\) and \(A_{12}^0\) have interference terms.

At the High Flux Reactor in Petten experiments on \(\gamma\)-emission after capture of polarized neutrons by polarized \(^{59}\text{Co}\) nuclei have been carried out [18].

The measured quantity is:

\[
R(\theta) = \frac{W(\theta)^{11} - W(\theta)^{11}}{W(\theta)^{11} + W(\theta)^{11}}
\]

for two directions \(\theta = 0^\circ\) and \(\theta = 90^\circ\). In this way information about \(A_{11}^0\) and \(A_{22}^0\) can be obtained.

Anisotropy measurements [19] with unpolarized neutrons give information about \(A_{22}^0\). In addition a circular polarization experiment [20] with polarized neutrons on unoriented \(^{59}\text{Co}\) has been carried out in Petten. In this way spin assignments for a number of \(^{60}\text{Co}\) levels have been obtained; in fact they were over-determined. The internal consistency of these 3 measurements was excellent. Interference effects were clearly demonstrated in this case.

Using a strong superconducting split solenoid to polarize nuclei, it will be possible to apply this type of experiment to many stable isotopes.

5. Fission of aligned nuclei by slow-neutron capture. — As a second example I like to consider a series of experiments concerning fission of aligned heavy nuclei by low energy neutrons generated with the aid of an electron linear accelerator at Harwell. The emission of fission fragments from aligned \(^{233}\text{U}\), \(^{235}\text{U}\) and \(^{237}\text{Np}\) as a function of neutron energy has been studied [20]-[22]. Alignment of the uranium isotopes was achieved through electric quadrupole coupling in the \(\text{UO}_2\) group of single crystals of \(\text{RbUO}_3(\text{NO}_3)_3\). In the case of \(^{237}\text{Np}\) a mixture of magnetic and electric hyperfine interactions occurs. It was necessary to detect the fission fragments inside the cryostat with Si-detectors mounted onto a 1 K shield.

The angular distribution of the fission fragments depends on the relative orientation of the deformation axis with respect to the nuclear orientation axis when the scission point is approached. The essential information to be obtained from this experiment concerns the projection quantum number, \(K\), of the compound spin, \(J\), with respect to the deformation axis. \(K\) is a characteristic parameter for the fission channels. The angular distribution of the fission fragments is given by (neglecting high-order terms):

\[
W(\theta) = 1 + \frac{15}{4 I + 1} \times f_2(I) \times \frac{3 K^2 - J(J + 1)}{J(J + 1)} P_2(\cos \theta).
\]

It is clear that the angular distribution depends very strongly on \(K\).

The most important conclusions from these experiments are, that \(a\) fission of \(^{233}\text{U}\) and \(^{235}\text{U}\) occurs through channels of low \(K\)-values and \(b\) that \(K\) is large for fission of \(^{237}\text{Np}\) (with \(J = 3\ K = 3\ or 2\) and with \(J = 2\ K = 2\)). It should be remarked that for these experiments counting periods of several months were necessary. This could only be achieved with a dilution refrigerator. A problem was the \(\alpha\)-activity of the samples, which generated large heat inputs and produced radiation damage of the detectors.

6. Hyperfine information from nuclear orientation experiments. — Nuclear orientation can be useful for obtaining information about internal magnetic
fields and electric field gradients at radioactive nuclei in solid state materials providing that sufficient information about the nuclear physics involved is known. The nuclear magnetic dipole and/or electric quadrupole moments should be known. If angular distributions of radioactive nuclei are used it is necessary to know the decay scheme sufficiently well. A systematic knowledge of hyperfine data of nuclei in solid state materials is of great interest to understand the interaction of ions with the surrounding material. It may also be possible to gain some insight about the composition of the lattice and positions of impurity atom and possible also vacancies from such data in combination with other experimental information.

In principle, it is possible to obtain both the internal magnetic field and electric field gradient at the nucleus if a sufficiently accurate measurement of the temperature dependence of the nuclear orientation effect is carried out. In most cases, however, one of the two terms is mainly determining the orientation behaviour. Consequently the obtainable information is usually limited. As an example the degree of nuclear polarization of holmium deduced from the transmission of polarized neutrons by polarized nuclei has been used to determine the magnetic field at the nucleus. It was not possible to derive the electric quadrupole interaction term in this case [24].

In cases of implanted ions in ferromagnetic metals it often occurs, that not all the nuclei feel the same internal magnetic field. This modifies the temperature dependence of the nuclear orientation parameters. If all the radioactive nuclei feel magnetic fields, not necessary of the same magnitude, the maximum possible anisotropy will ultimately be reached at sufficiently low temperatures. The orientation parameters are weighted averages over the various sub-ensembles of nuclei feeling different fields; that is \( f_k = \frac{\sum f_k(i) w(i)}{\sum w(i)} \), where \( w(i) \) are weighing factors with the normalization condition \( \sum w(i) = 1 \). Usually it will not be possible to analyse in this way the observed anisotropy as a function of temperature if more than two sub-ensembles with different fields are assumed. In the simple case of a sample in which a fraction \( w \) of the nuclei feels a negligibly small magnetic field the anisotropy is reduced by a factor \( 1 - w \). Work on Xe isotopes carried out in Louvain by Silverans and others [25] and on I isotopes in Groningen by Koene [26] have shown that the amount of radioactive nuclei implanted in iron, which feel a small field, can be estimated fairly accurately from the observed anisotropies. Mössbauer experiments enable to obtain more detailed information about the different fractions since they show up as different overlapping Mössbauer spectra [27].

An interesting extension to nuclear orientation is obtained by introducing nuclear magnetic resonance techniques. By varying the frequency of a radiofrequency field applied perpendicular to the magnetizing field, it is possible to destroy or at least to reduce the nuclear orientation. At the resonance frequency \( \nu \) we have the relation \( h\nu = \mu H/I \) if only magnetic dipole interaction is considered. If different internal magnetic fields exist different resonances will be found. An interesting example of NMR on oriented nuclei is given by Bacon et al. [28] who applied this technique on \(^{206}\text{Pb}\) in nickel foils. By measuring the anisotropy as a function of temperature and by determining the resonance frequency both \( \mu H \) and \( \mu H/I \) were obtained. Since the magnetic field of gold isotopes in nickel is well known, they were able to derive \( \mu \) and \( I \) of this isomer.

7. Concluding remarks. — In this talk the development of static nuclear orientation methods and their applications in nuclear physics have been briefly reviewed. I like to finish this talk with a few general remarks:

a) Nuclear orientation can now be used in the study of the decay of many radioactive isotopes if they have sufficiently long half lives. Also prompt radioactivity can be studied if the heat input is low as is the case with neutron beams. It would be of great interest to develop techniques to introduce nuclei of shorter half lives inside a working dilution refrigerator. This may enable the study of isotopes with half lives of the order of one hour.

b) Experiments with neutron beams and oriented nuclei are well feasible. Beams of charged particles introduce a large amount of heat and are therefore a problem. However, if low beam intensities are acceptable several experiments with oriented nuclei seem feasible.

c) A further development of techniques relevant to nuclear orientation can be expected, thus increasing the possibilities of nuclear orientation still further.

d) Nuclear orientation techniques are likely to remain difficult. They should therefore be restricted to cases where other experiments are not able to give reliable or sufficiently complete information. Nuclear orientation is a field in which nuclear, solid-state and low-temperature physicists can find a challenging cooperation.

References


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