# Direct Spin Determination of On-Line-Separated Isotopes by Nuclear Orientation and Nuclear Magnetic Resonance

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Nuclear magnetic resonance of on-line-oriented nuclei is reported for the first time, in the cases of mass-separated $^{106}$In, $^{108}$In, and $^{104}$Ag. Combination of the extracted $g$ factors with independent information on the magnetic-moment values leads to unambiguous spin assignment for the high-spin ground states of $^{106}$In and $^{108}$In. The high-precision magnetic moments derived for these nuclei as well as the result on $^{104}$Ag point to strong discrepancies from the additivity relation.

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Unambiguous spin assignment to ground states of nuclei is a first requirement for reliable nuclear-structure studies. Clearly for these ground-state spin determinations, only "strong arguments" may be used. This means that one should use *direct* measurements which are independent of decay-scheme considerations. Incomplete and/or erroneous decay schemes have often led to a time dependence of spin assignments based on weak arguments, as, e.g., in the intensively studied cases of $^{106}$In and $^{108}$In.[1-8]

Unfortunately, the choice of methods for ground-state spin determinations which are in accordance with on-line separation conditions is rather limited. The atomic-beam method, which is the ideal method, suffers from low transmission rates, severely restricting its application far from stability.[9]

Alternative methods rely on the *resolution* of the hyperfine structure. This approach may be used, for instance, in laser spectroscopy or in nuclear quadrupole resonance (NQR) spectroscopy. At high spin, though, the hyperfine patterns become highly complicated and overlapping. A clear spin assignment is obtainable when combining laser spectroscopy with rf, but then intensity problems may arise again. This particular technique is being tested for on-line use by at least one group.[10] The principle of NQR has been applied in a quadrupole-interaction nuclear magnetic resonance on oriented nuclei (NMR-ON) experiment on $^{188}$Ir,[11] but the method remains restricted to a few favorable cases.

A more generally applicable direct method has been developed recently at the nuclear orientation on-line facility KOOL, where nuclear orientation of almost all isotopes produced by the combination of the CYCLONE cyclotron and the LISOL on-line separator can be achieved.[12] The NMR-ON technique, however, was applied up to now only to long-lived isotopes—a review is given by Herzog.[13] The need for high statistics and long search times made this technique to be considered incompatible with on-line working conditions. We succeeded in obtaining NMR-ON curves for $^{106}$In, $^{108}$In, $^{104}$Ag, and $^{189}$Au$^{m}$. The last case concerned the study of the high field fraction of implantation sites.[14]

An NMR coil was installed around the continuously implanted sample, which was connected to the mixing chamber of a $^{3}$He-$^{4}$He dilution refrigerator. The equipment and the acquisition method were developed and tested for on-line use. Special care was taken to eliminate the effects of varying production rates by taking the ratio $W(0)/W(\pi/2)$ as the anisotropy function, where $W(\theta)$ stands for the emission probability at angle $\theta$. Moreover, the temperature was continuously monitored for the presence of rf-power-induced pseudoresonances. By alternating counting periods with unmodulated and modulated rf we obtained an adequate normalization for each point separately, and sufficient time was left between subsequent counting periods to eliminate effects of relaxation rate. Each frequency region was scanned in both directions several times.

The data for $^{106}$In were obtained by use of a modulation sweep of 375 kHz and a frequency step of 500 kHz, at temperatures as low as 12 mK in conditions of continuous cold implantation and rf irradiation. The resonance curve obtained for the 633-keV transition in $^{106}$Cd is displayed in Fig. 1. A Gaussian fit leads to a center frequency $\nu_0 = 153.1(3)$ MHz while the FWHM is 2.4 MHz. Although the resonance curve cannot be strictly Gaussian [modulation width, combination of $W(0)$ and $W(\pi/2)$], our theoretical simulations show relatively small deviations for modulation sweeps less than $\sim 35\%$ of the line width. The integrated destruction is then calculated as 24%. Depending on whether we accept a spin value of 6 or 7, the corresponding $|\mu H|$ values are $120.5\mu_N$ T, or $140.6\mu_N$ T, respectively. Now we use these numbers to fit the anisotropy measurement of the 998-keV $6^+ \rightarrow 4^+$ $E2$ $\gamma$ transition in $^{106}$Cd, which is a firmly established yrast transition.[3,15] For spin 6 the closest fit assumes a *fully substitutional* implantation, and shows a clear discrepancy with the data. On the contrary, the excellent fit to the experimental points we obtain for spin 7, shows this value to be the only one compatible with the data (Fig. 2).

Recently another approach has become possible.

 

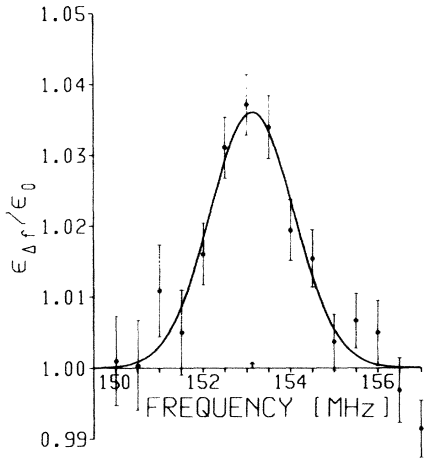


FIG. 1. Data points of the 633-keV $\gamma$ transition and associated Gaussian fit for the NMR-ON experiment on $^{106}$In. $\varepsilon_{\Delta f}$ and $\varepsilon_0$ stand for, respectively, the anisotropy recorded with and without frequency modulation of the rf signal.

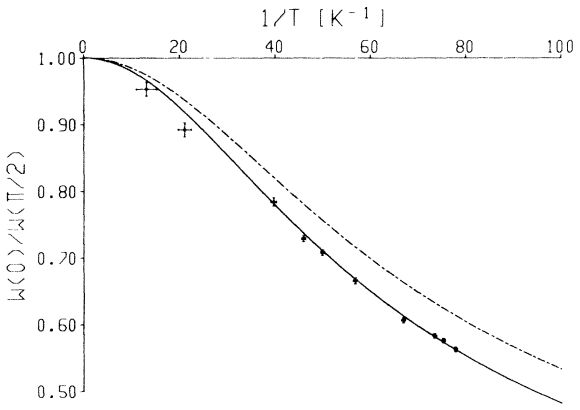


FIG. 2. Comparison of the closest fits for parent spin 7 (full line), and 6 (broken line) to the experimental anisotropies of the $6^+ \rightarrow 4^+$ $E2$ transition of 998 keV in the decay of $^{106}$In. The curve for spin 6 is drawn for 100% substitutionality.

From laser spectroscopy measurements the hyperfine splitting of the $6s\ ^2S_{1/2}$ state in $^{106}$In has been obtained, yielding $\mu(^{106}\text{In}) = 4.916(7)\mu_N$ (where $\mu_N$ is the nuclear magneton) for spin 7, and $4.862(7)\mu_N$ for spin 6, neglecting any hyperfine anomaly.[16] Under the same assumption, and using a hyperfine field of $-28.66(5)$ T,[17] we obtain $\mu(^{106}\text{In}) = 4.921(13)\mu_N$ or $4.218(11)\mu_N$ for spin 7 and 6, respectively. The spin value 7 is confirmed by the perfect agreement between the two corresponding moments.

In the case of $^{108}$In this approach is possible again: The moment values are available from recent laser spectroscopy as $4.513(3)\mu_N$ or $4.561(3)\mu_N$ for spin 6 and 7, respectively.[18,19] However, these hyperfine structure data do not allow a spin determination. The corresponding NMR-ON frequencies are 163.8(3) and 141.9(3) MHz. The result of the measurement (modulation width 2 MHz, step 2 MHz) is shown in Fig. 3. The fitted curve, with the fixed center frequency of 141.9 MHz and FWHM of 2.4 MHz, indicates an integrated destruction of 15%. Thus, once again, the spin value 7 is firmly established.

The Brennan-Bernstein rules,[20] as well as the Paar parabola rule,[21] predict that a $[\pi(g_{9/2})^{-1}\nu(d_{5/2})^{-1}]$ configuration gives rise to a $2^+$, $7^+$ ground-state multiplet. For a hole-particle coupling, on the other hand, the parabola rule gives a preferential ground-state spin 5. A nearly degenerate $2^+$, $7^+$ multiplet has been confirmed in the case of $^{110}$In. For $^{108}$In the high spin value 7 is now established, and an unambiguous low spin value of 2 is claimed by a "note added in proof" in Ref. 19. In $^{106}$In, a similar configuration is clearly suggested by the spin-7 result, and a low spin value of 2 becomes highly probable, too.

In general the validity of the additivity relation for magnetic moments is remarkable, in spite of its empirical character in real nuclei, where configuration mixing and residual interactions are important. For the three nuclei of interest a comparison is listed in Table I. By use of the neighboring moments (known with high accuracy[22]), the empirical values are given for both spin $J_\nu = \frac{5}{2}$ and $J_\nu = \frac{7}{2}$, since an important mixing between the $\nu(2d_{5/2})$-based and the $\nu(1g_{7/2})$-based $7^+$ configurations is expected. The rather isolated $\pi(1g_{9/2})$ orbital and the constancy of the magnetic moments in the chain of the odd In isotopes from mass 127 down to 105 suggest a unique proton configuration. It is a surprise to note that, in $^{108}$In, a severe discrepancy of $\sim 10\%$ between the experimental and the empirical values for the $J_\nu = \frac{5}{2}$ occurs,

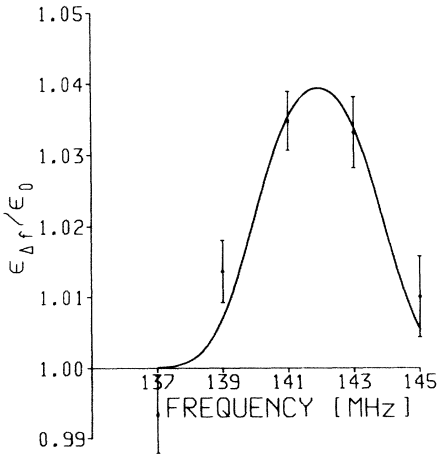


FIG. 3. Data points of the 633-keV $\gamma$ transition from the NMR-ON experiment on $^{108}$In. $\varepsilon_{\Delta f}$ and $\varepsilon_0$ stand for the anisotropy recorded, respectively, with and without frequency modulation of the rf signal. The full curve gives the calculated curve as expected for spin 7 (center = 141.9 MHz, intrinsic width = 2.4 MHz).

TABLE I. Comparison of the experimental ($\mu_{expt}$), empirical ($\mu_{emp}$) and theoretical ($\mu_{th}$) magnetic-moment values for the high-spin ground states in $^{106}$In, $^{108}$In, and $^{110}$In. The theoretical values are taken from Ref. 23.

| Mass number | $I^\pi$ | $\mu_{expt}$ ($\mu_N$) | $J_\nu$ | $\mu_{emp}$ Moment ($\mu_N$) | $\mu_{th}$ ($\mu_N$) |
|---|---|---|---|---|---|
| 106 | $7^+$ | 4.921(13)[a] | $\frac{5}{2}$ | 4.975 | 4.415 |
| | | | $\frac{7}{2}$ | 5.53 | |
| 108 | $7^+$ | 4.561(3)[b] | $\frac{5}{2}$ | 4.291 | 5.40 |
| | | | $\frac{7}{2}$ | 5.53 | |
| 110 | $7^+$ | 4.719(13)[c] | $\frac{5}{2}$ | 4.711 | 4.876 |

[a]This work.
[b]References 18, 19, and this work.
[c]Reference 17.

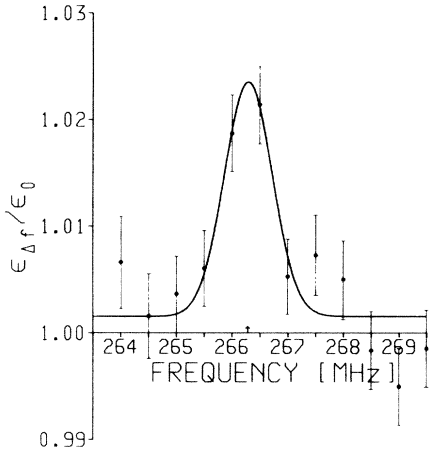


FIG. 4. Data points of the 768-keV $\gamma$ transition and associated Gaussian fit for the NMR-ON experiment on $^{104}$Ag. $\varepsilon_{\Delta f}$ and $\varepsilon_0$ stand for, respectively, the anisotropy recorded with and without frequency modulation of the rf signal.

while the correspondence is totally satisfactory for $^{110}$In and for $^{106}$In. Such a deviation has to be explained by the collective aspects of the wave function, which clearly violate the additivity rule. It shows that in In nuclei the empirical additivity rule cannot be expected to hold and cannot be used as a basis for the characterization of the ground-state configuration(s). Instead, the interpretation should rely on a detailed calculation of the ground-state wave functions.

Therefore, we include recent theoretical results[23] from a calculation in the framework of a neutron quasiparticle proton-hole coupling to the quadrupole vibrations of the core. The merit of these results should be evaluated in their possibility of reproducing trends rather than absolute values since effective $g_p$ and $g_n$ factors have been used (e.g., half of the free-particle value for the spin part of $g$).

In our case, theory does not describe the moment systematics adequately. The low experimental value for $^{108}$In is not reproduced, and the very strong deviation in this case (much larger than the reliability of the calculation, i.e., 5%–10%) gives a good indication that the importance of the $g_{7/2}$ orbital has been overestimated. A determination of the spin and the moment value of the $^{104}$In ground state might help to clarify the situation by indicating whether a "staggering" of the even In moments really exists. These measurements are in progress.

The breakdown of the additivity rule in In prompted us to look for similar effects in neighboring nuclei, especially Ag. In $^{104}$Ag ($Z=47$, $N=57$) the previously reported moment value of $\mu=(4.0^{+0.2}_{-0.1})\mu_N$[24] suggested another possible deviation. The lowest states of the odd-odd Ag isotopes are usually described as $[\pi(g_{9/2})_{J_\pi}^{-3}\text{-}\nu(d_{5/2})^{-1}]_{I^+}$, with $J_\pi=\frac{7}{2}$ (anomalous coupling), $I=6$ for the high-spin isomers in $^{106\text{-}110}$Ag. Their $g$ factors are properly reproduced by the additivity relation. The $I^\pi=5^+$ ground state in $^{104}$Ag[25] may be considered as a mixing of both the seniority $v=1$ ($J_\pi=\frac{9}{2}$) and seniority $v=3$ ($J_\pi=\frac{7}{2}$) configurations. Empirical $g$ factors of 0.672(9) and 0.906(11) are deduced for the two "pure" couplings, built on a $\frac{7}{2}^+$ and $\frac{9}{2}^+$ cluster, respectively. An accurate moment determination may certainly help to characterize the ground state.

NMR-ON was searched for in the relevant frequency region with a step and width identical to those used in the case of $^{106}$In. The resonance curve for the 768-keV transition in $^{104}$Pd is shown in Fig. 4. A resonance frequency of 266.3(5) MHz and FWHM of 1.3 MHz can be deduced from a Gaussian fit. By use of the hyperfine field of $-44.72(2)$ T,[26] a $g_{expt}=0.7828(16)$ is obtained. This experimental value clearly deviates from both empirical extrapolations, supporting the suggestion of an important mixing of the configurations based on the two proposed proton clusters.

We have been able to demonstrate that the on-line application of the nuclear orientation method in combination with nuclear magnetic resonance provides a powerful technique for directly determining ground or isomeric state spins of short-lived isotopes. It is important to note that the high resolving power of this method enables one to distinguish between high spin values. Furthermore, *precise* magnetic-moment values of nuclei far from stability provide basic test results for model descriptions, and the systematics should be extended towards the neutron shell closure. As a final remark we draw attention to the fact that the combination of NMR-ON with independent information on the nuclear moments, especially from laser spectroscopy, yields a fast and efficient spin determination. Mainly for high-spin nuclei, the two methods complement each other perfectly.

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[1]S. Flanagan, R. Chapman, J. L. Durell, W. Gelletly, and J. N. Mo, J. Phys. G **2**, 589 (1976).

[2]H. Huang, B. P. Pathak, and J. K. P. Lee, Can. J. Phys. **56**, 936 (1978).

[3]L. E. Samuelson, J. A. Grau, S. I. Popik, F. A. Rickey, and P. C. Simms, Phys. Rev. C **19**, 73 (1979).

[4]I. N. Wischnewski, H. V. Klapdor, P. Herges, H. Fromm, and W. A. Zheldonozhski, Z. Phys. A **298**, 21 (1980).

[5]B. Roussière, P. Kilcher, J. Sauvage-Letessier, C. Bourgeois, R. Béraud, R. Duffait, M. Meyer, J. Genevey-Rivier, and J. Tréherne, Nucl. Phys. **A419**, 61 (1984).

[6]S. Flanagan, R. Chapman, G. D. Dracoulis, J. L. Durell, W. Gelletly, A. J. Hartley, and J. N. Mo, J. Phys. G **1**, 77 (1975).

[7]L. E. Samuelson, F. A. Rickey, J. A. Grau, S. I. Popik, and P. C. Simms, Nucl. Phys. **A301**, 159 (1978).

[8]N. Elias, Ph.D. thesis, Université de Lyon, 1979 (unpublished).

[9]C. Ekström, S. Ingelman, and G. Wannberg, Nucl. Instrum. Methods **148**, 17 (1978).

[10]N. Bendali, H. T. Duong, J. M. Saint-Jalm, and J. L. Vialle, J. Phys. (Paris) **45**, 421 (1984).

[11]R. Eder, E. Hagn, and E. Zech, Phys. Rev. C **32**, 582 (1985).

[12]D. Vandeplassche, L. Vanneste, H. Pattyn, J. Geenen, C. Nuytten, and E. van Walle, Nucl. Instrum. Methods **186**, 211 (1981).

[13]P. Herzog, in *Nuclear Orientation,* edited by H. Postma and N. J. Stone (North-Holland, Amsterdam, 1986).

[14]E. van Walle, D. Vandeplassche, J. Wouters, N. Severijns, and L. Vanneste, Phys. Rev. B **34**, 2014 (1986).

[15]D. C. Stromswold, D. O. Elliott, Y. K. Lee, L. E. Samuelson, J. A. Grau, F. A. Rickey, and P. C. Simms, Phys. Rev. C **17**, 143 (1978).

[16]H. Lochmann, U. Dinger, J. Eberz, G. Huber, R. Menges, G. Ulm, R. Kirchner, O. Klepper, T. U. Kühl, D. Marx, and D. Schardt, Z. Phys. A **322**, 703 (1985).

[17]E. Hagn, E. Zech, and G. Eska, Z. Phys. A **300**, 339 (1981).

[18]G. Ulm, J. Eberz, G. Huber, H. Lochmann, R. Menges, R. Kirchner, O. Klepper, T. Kühl, P. O. Larsson, D. Marx, D. Murnick, and D. Schardt, Z. Phys. A **321**, 395 (1985).

[19]J. Eberz, U. Dinger, T. Horiguchi, G. Huber, H. Lochmann, R. Menges, R. Kirchner, O. Klepper, T. Kühl, D. Marx, E. Roeckl, D. Schardt, and G. Ulm, Z. Phys. A **323**, 119 (1986).

[20]M. H. Brennan and A. M. Bernstein, Phys. Rev. **120**, 927 (1960).

[21]V. Paar, Nucl. Phys. **A331**, 16 (1979).

[22]*Table of Nuclear Moments,* edited by V. S. Shirley and C. M. Lederer (Wiley, New York, 1978), Appendix VII, pp. 42–64.

[23]J. Van Maldeghem, K. Heyde, and J. Sau, Phys. Rev. C **32**, 1067 (1985).

[24]O. Ames, A. M. Bernstein, M. H. Brennan, and D. R. Hamilton, Phys. Rev. **123**, 1793 (1961).

[25]O. Ames, A. M. Bernstein, M. H. Brennan, R. A. Haberstroh, and D. R. Hamilton, Phys. Rev. **118**, 1599 (1960).

[26]R. A. Fox, P. D. Johnston, and N. J. Stone, Phys. Lett. **34A**, 211 (1971).