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Nuclear magnetic moment of ^{69}As from on-line β-NMR on oriented nuclei

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A precise value for the magnetic moment of the ^{69}As $5/2^-$ ground state has been obtained from nuclear magnetic resonance on oriented nuclei (NMR/ON) using the NICOLE ^{3}He-^{4}He dilution refrigerator setup at ISOLDE/CERN. The NMR/ON signal was observed by monitoring the anisotropy of the ^{69}As β particles. The center frequency $\nu[B_{\text{ext}} = 0.0994(10)T] = 169.98(9)$ MHz corresponds to $\mu[^{69}\text{As}] = +1.6229(16)\mu_N$. This result differs considerably from the $\pi f_{5/2}$ single-particle value obtained with g factors for a free proton but is in reasonable agreement with the value obtained with effective g factors and with values from a core polarization calculation and from calculations in the framework of the interacting boson-fermion model. Assuming a single exponential spin-lattice relaxation behavior a relaxation time $T_1' = 10(25)$ s was observed for ^{69}As$\underline{\text{Fe}}$ at a temperature of about 20 mK in a magnetic field $B = 0.1$ T.

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I. INTRODUCTION

Nuclei in the $A = 70$ mass region exhibit a large variety of complex phenomena, including, e.g., large deformations, shape transitions, and shape coexistence. The description of these nuclei is complicated because of the large number of nucleons outside the closed neutron and proton shells $N = Z = 28$, which leads to a variety of possible configurations. Also, because of the presence of several large gaps in the Nilsson scheme for both protons and neutrons in this mass region, the nuclear structure often changes drastically by adding just a few nucleons. All these aspects make the $A = 70$ mass region an interesting testing ground for different theoretical approaches [1–4].

Precise measurements of magnetic moments of nuclei can help to determine which of various possible configurations is realized in a certain isotope, thus providing basic tests of nuclear models. The combination of low temperature nuclear orientation (LTNO) with nuclear magnetic resonance on oriented nuclei (NMR/ON) [5] is well suited to determine the magnetic moment of ground states with good precision. Here we present a β-NMR/ON measurement of the magnetic moment of ^{69}As ($t_{1/2} = 15.23$ min, $I^\pi = 5/2^-$) with much improved accuracy compared to the previously published results, viz. $\mu = 1.2(2)\mu_N$ [6] and $\mu = +1.58(16)\mu_N$ [7] (see also Ref. [8]).

II. EXPERIMENT AND FORMALISM

A 1.4-GeV pulsed proton beam (3×10^{13} protons per pulse) from the CERN Proton Synchrotron Booster accelerator (PS Booster) was used to produce the ^{69}As nuclei by spallation in a 11 g/cm^2 ZrO_2 fiber target [9]. The reaction products diffused out of the 1900°C hot target and effused via a hot transfer line to a FEBIAD-type "hot plasma" ion source. The atoms were ionized to 1^+ ions, extracted, accelerated to 60 keV, mass separated by the ISOLDE General Purpose Separator (GPS) [10,11], and finally transported through the beam distribution system to be implanted into an iron foil inside the NICOLE ^{3}He-^{4}He dilution refrigerator [8,12]. The ^{69}As$^+$ beam intensity after mass separation was typically about 2×10^6 ions/s at an average proton beam intensity of 2.5 μA. The iron implantation foil had been prepared by polishing a 99.99% pure Fe foil (thickness 250 μm) from Goodfellow and then annealing it at 800°C for 6 hr in a hydrogen atmosphere. This sample foil was then soldered with Woods metal to the sample holder of the dilution refrigerator.

The angular distribution of the β particles emitted by the implanted ^{69}As nuclei was observed with HPGe particle detectors [13,14] with a sensitive area of about 110 mm^2, mounted inside the 4K radiation shield of the refrigerator at a distance of about 32 mm from the sample. Operating these detectors inside the helium cooled shield, i.e., without any window between source and detector, avoids energy losses and reduces scattering of the β particles. In addition, the thickness of the detectors was chosen such that they could just fully stop the end-point energy β particles of ^{69}As, thereby keeping the sensitivity to γ rays and thus the γ contamination in the β spectra to a minimum. Moreover, with γ absorption in the particle detectors being minimal the detection of γ radiation by the large-volume HPGe detectors installed outside the refrigerator was not hindered too much. The detectors were mounted slightly tilted (i.e., at an angle of about 15°) with respect to the magnetization axis to minimize the influence of scattering effects in the Fe host foil. Thin isolated copper wires (about 13 cm long) connected the detectors to the preamplifiers that were placed outside the refrigerator, resulting in an energy resolution of about 3 keV for 1-MeV β particles.

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Angular distributions of γ rays were observed as well. For this, three large volume HPGe detectors with an energy resolution of about 3 keV at 1332 keV were used. These were placed outside the refrigerator, two along the polarization axis and one perpendicular to it. They mainly served to monitor the temperature of the sample by observing the anisotropy of the 136-keV γ ray from a calibrated $^{57}\text{Co}\underline{\text{Fe}}$ nuclear orientation thermometer [15].

The angular distribution of radiation emitted from an axially symmetric ensemble of oriented nuclei is given by [16] the following:

$$W(\theta) = 1 + f \sum_{\lambda} B_{\lambda}(\mu B/k_B T, I)\, U_{\lambda} A_{\lambda} Q_{\lambda} P_{\lambda}(\cos\theta), \quad (1)$$

where f represents the fraction of the nuclei that experience the full hyperfine interaction, whereas the rest $(1-f)$ is supposed to feel no interaction at all; the B_{λ} describe the nuclear orientation and depend on the magnetic moment μ of the decaying nuclei, the total magnetic field B the nuclei experience (i.e., the sum of the hyperfine magnetic field B_{hf}, the applied field B_{applied}, and the demagnetization field B_{dem}), the temperature of the sample T, and the initial spin I of the oriented state; the U_{λ} are the deorientation coefficients that account for the effect of unobserved intermediate radiations; the A_{λ} are the directional distribution coefficients that depend on the properties of the observed radiation and the nuclear levels involved; the Q_{λ} are solid angle corrections factors; and $P_{\lambda}(\cos\theta)$ are the Legendre polynomials. The detection angle θ is measured relative to the direction of the saturation magnetization axis of the Fe host foil, which is defined by the applied magnetic field.

For γ rays only λ even terms occur. For positrons from allowed β decays only the $\lambda = 1$ term is present and Eq. (1) transforms to the following:

$$W(\theta) = 1 + f\frac{v}{c} B_1 A_1 Q_1 \cos\theta, \quad (2)$$

where v/c is the electron velocity relative to the speed of light.

The experimental angular distribution is given by the following:

$$W(\theta) = \frac{N_{\text{cold}}(\theta)}{N_{\text{warm}}(\theta)}, \quad (3)$$

with $N_{\text{cold,warm}}(\theta)$ the count rates when the sample is cold (about 10 mK; polarized nuclei) or warm (about 1K; unpolarized nuclei). In on-line experiments, where the count rates vary with beam intensity, it is customary to construct a double ratio, combining count rates in two different detectors. In the present work this double ratio was

$$R = \left(\frac{N(165^\circ)}{N(15^\circ)}\right)_{\text{cold}} \Bigg/ \left(\frac{N(165^\circ)}{N(15^\circ)}\right)_{\text{warm}}. \quad (4)$$

All data were corrected for the dead time of the data acquisition system using a precision pulse generator.

For the NMR/ON experiment an RF oscillating field was applied perpendicular to the external magnetic field. The NMR coil producing this oscillating field consisted of a pair of two-turn coils mounted on a Teflon frame that was fixed inside the radiation shield around the sample. The coil was fed from the top of the refrigerator by coaxial cables connected to a Marconi frequency generator with a range from 10 kHz to 3.3 GHz. In addition to the NMR coil a pickup coil for monitoring the RF signal was present too. The intensity of the RF signal was kept as small as possible to avoid a too large RF heating and subsequent reduction in anisotropy for the β particles.

The resonance measurement was performed by scanning the radio frequency ν and observing the destruction of the anisotropy of the β radiation from the nuclei. At the resonance frequency ν_{res} transitions between the Zeeman splitted nuclear sublevels are induced, thus reducing the degree of nuclear polarization and therefore the magnitude of the anisotropy $R-1$. The resonance frequency is related to the nuclear magnetic moment through the relation

$$\nu_{\text{res}} = \left|\frac{\mu}{Ih}[B_{\text{hf}} + B_{\text{applied}} - B_{\text{dem}}]\right|. \quad (5)$$

For the 250-μm-thick Fe foil that was used here a demagnetization field $B_{\text{dem}} = 0.038(8)$ T was calculated [17]. To fully saturate the iron foil, first an external magnetic field of 0.5 T was applied, which was later reduced to $B_{\text{applied}} = 0.0994(10)$ T to minimize its influence on the trajectories of the β particles. The hyperfine field of As in Fe is $B_{\text{hf}} = +34.29(3)$ T [18].

The β decay of ^{69}As proceeds mainly through two branches [19], the most intense one being the allowed $5/2^- \to 5/2^-$ ground state to ground-state Gamow-Teller transition with end-point energy $E_0 = 2991$ keV and a β^+ intensity of 73%. The second most intense branch, with a β^+ intensity of 14%, feeds the $3/2^-$ second excited level of ^{69}Ge (at 233 keV). The rest of the β^+ intensity is spread over various much weaker branches. Because in a β-NMR/ON experiment one is interested in the destruction of the asymmetry in the angular distribution pattern of the β particles, the entire energy region from 1205 keV (the β-spectrum end-point energy of ^{69}Ge, the daughter isotope of ^{69}As) to 2991 keV was used to improve statistics (Fig. 1). This was possible, even though the β^+ anisotropy depends on the energy of the β particles [see Eq. (2)], because the positron velocity relative to the speed of light (v/c) changes only from 0.95 to 0.99 in this energy region, whereas, in addition, the β anisotropy was, within error bars, found to be the same throughout this energy region.

From the γ spectrum it was deduced that there was also a small ^{69}Cu component in the beam. From the intensity of the ^{69}Cu γ rays, corrected for the detector efficiency, the amount of ^{69}Cu relative to ^{69}As was found to be 28(5)%. Taking then into account the β branching ratios for both isotopes in the energy region from 1205 to 2675 keV (the β end-point energy for ^{69}Cu), the amount of counts coming from ^{69}Cu in this energy region of the β spectrum was calculated to be 14(3)%. Because ^{69}Cu and ^{69}As have different hyperfine interaction frequencies, the effect of this modest contamination was to slightly reduce the sensitivity of the resonance experiment.

For a relatively short lived nucleus with a moderate hyperfine interaction strength $T_{\text{int}} = |\mu B/k_B I|$, with k_B the Boltzmann constant, the observed anisotropy can be reduced from its maximal value (which corresponds to thermal equilibrium) because of incomplete spin-lattice relaxation (i.e.,

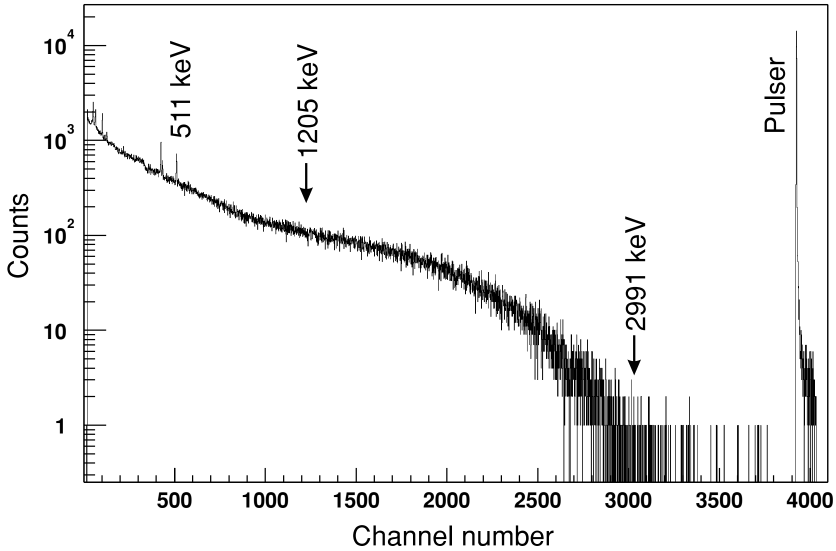

FIG. 1. Typical β spectrum recorded within one 300 s measurement cycle. The 511-keV positron annihilation line and the pulser peak are indicated. The spectrum contains mainly counts from β particles of ^{69}As (β end-point energy $E_0 = 2991$ keV), but also from ^{69}Cu ($E_0 = 2675$ keV) and ^{69}Ge ($E_0 = 1205$ keV). The part of the spectrum between 1205 and 2991 keV was used for analysis (see text).

Refs. [20,21]). The spin-lattice relaxation rate is determined by the so-called Korringa relaxation constant C_K, which is related to T_{int}, the sample temperature T_L, and the relaxation time T_1' (assuming a single exponential relaxation behavior) by [20] the following:

$$T_1' \approx \begin{cases} C_K/IT_{\text{int}} & \text{for } T_L \leqslant IT_{\text{int}} \\ C_K/T_L & \text{for } T_L \geqslant IT_{\text{int}}. \end{cases} \tag{6}$$

With $I = 5/2$ and $T_{\text{int}} = 8$ mK for the ground state of ^{69}As$\underline{\text{Fe}}$, the empirical relation for the relaxation of impurities in Fe in a magnetic field $B \geqslant 1$ T, viz. [20]

$$C_K T_{\text{int}} = 1.4 \times 10^{-4} \text{ s K}^3 \tag{7}$$

(which is usually correct within a factor of 4), yields $C_K \approx 2200$ s mK. Taking into account that in a field of about 0.1 T, as was used here, spin-lattice relaxation was found to proceed about 2 to 3 times faster [20], a spin-lattice relaxation time T_1' of several tens of seconds is expected for ^{69}As in Fe at temperatures T_L of about 20 mK at which the resonance experiment was performed. This would cause only a small reduction of the β anisotropy compared to the case of full relaxation (i.e., when $T_1' = 0$) [21].

III. DATA COLLECTION AND ANALYSIS

First, the magnetic moment of ^{69}As was estimated from the temperature dependence of the β anisotropy observed in a classical LTNO experiment that was carried out prior to the NMR/ON measurements. This yielded $\mu = +1.52(19)\mu_N$, which is in agreement with and of similar precision as the result from a previous LTNO experiment, viz. $\mu = +1.58(16)\mu_N$ [7,8]. These values were then used to establish the, albeit rather broad, search region for the NMR/ON experiment.

In the NMR/ON experiment about 200 spectra of 300 s each were recorded. During the measurements the sample temperature varied between 15 and 22 mK because of the different eddy current heating response of the system at different frequencies. First, two downward scans, followed by two upward scans, were carried out in the region between 160 and 180 MHz. Frequency steps of 2 MHz were chosen with a 2.8-MHz modulation amplitude and a 100-Hz modulation

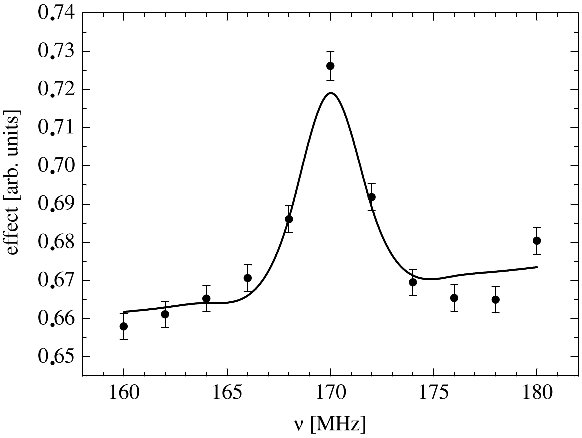

FIG. 2. On-line NMR/ON curve for ^{69}As obtained when scanning the frequency with steps of 2 MHz.

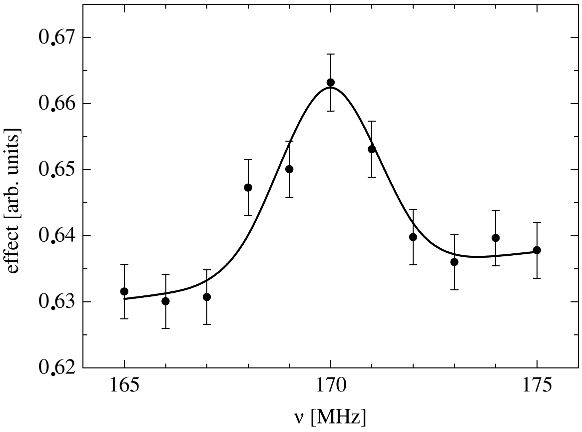

FIG. 3. On-line NMR/ON curve for ^{69}As obtained when scanning the frequency with steps of 1 MHz.

TABLE I. Experimental magnetic moments of the lowest $5/2^-$ state in the odd-A As isotopes compared with theoretical results obtained in the core polarization model of Noya, Arima, and Horie [25] for different proton and neutron configurations.

N	Isotope	$E_{5/2}$ (keV)	μ_{exp} (μ_N)	μ_{th} (μ_N)	Proton configuration	Neutron configuration
36	^{69}As	0	+1.6229(16)[a]	1.68[b]	$(2p_{3/2})^2(1g_{9/2})^2(1f_{5/2})^1$	$(1f_{5/2})^6(2p_{3/2})^2$
38	^{71}As	0	(+)1.6735(18)[c]	1.81[d]	$(2p_{3/2})^2(1g_{9/2})^2(1f_{5/2})^1$	$(1f_{5/2})^6(2p_{3/2})^4$
40	^{73}As	67	+1.63(10)[c]	1.72[d]	$(2p_{3/2})^2(1g_{9/2})^2(1f_{5/2})^1$	$(1f_{5/2})^6(2p_{1/2})^2$
42	^{75}As	280	+0.918(18)[e]	0.96[d]	$(1f_{5/2})^5$	$(2p_{3/2})^4(1g_{9/2})^4$
44	^{77}As	264	+0.736(22)[e]	0.91[d]	$(1f_{5/2})^5$	$(2p_{3/2})^4(2p_{1/2})^2(1g_{9/2})^4$

[a] This work; β-NMR/ON.

[b] Calculated in the core polarization model of Ref. [25] for the given proton and neutron configuration.

[c] Ref. [26].

[d] Core polarization calculations along Ref. [25] given in Ref. [26] for the given proton and neutron configuration.

[e] Ref. [27]; time-differential perturbed angular correlations.

frequency. A clear resonance was observed at a frequency of about 170 MHz. Then, two more detailed scans with frequency steps of 1 MHz, with a 1.1-MHz modulation amplitude and a 100-Hz modulation frequency, were performed in the region from 175 to 165 MHz.

All six scans were least-squares fitted simultaneously, allowing for a nonzero spin-lattice relaxation time T_1', yielding

$$\nu_{\text{res}} = 169.98(9)\text{MHz} \tag{8}$$

and

$$T_1' = 10(25)\,\text{s}. \tag{9}$$

The value obtained for T_1' is in rough agreement with expectations (see Sec. II). The data and fit results for two scans are shown in Figs. 2 and 3.

IV. RESULTS AND DISCUSSION

The resonance frequency $\nu_{\text{res}} = 169.98(9)$ MHz corresponds to a nuclear magnetic moment

$$\mu[^{69}\text{As}] = +1.6229 \pm 0.0016\,\mu_N. \tag{10}$$

The sign was obtained from the observed β asymmetry in the LTNO experiment.

The Schmidt (single particle) value for the $5/2^-$ ground state of the odd As isotopes is $\mu_{\text{sp}} = +0.86\,\mu_N$. However, using effective g factors $g_s^{\text{eff}}(\pi) = 0.7 g_s^{\text{free}} = +3.906$ and $g_l^{\text{eff}}(\pi) = +1.1$, leads to $\mu_{\text{sp}}^{\text{eff}} = +1.75\,\mu_N$, in good agreement with our result.

Because ^{69}As is a strongly deformed nucleus (deformation parameter $\beta_2 = 0.29$ [22]) the core also contributes to the magnetic moment. Using then the relation for a strong coupled system consisting of a single particle and a deformed core [23,24]

$$\mu_{\text{sc}} = \mu_{\text{sp}} - (g_j - g_R)\frac{I}{I+1}\mu_N, \tag{11}$$

with $g_R = Z/A$ the angular momentum carried by the core, and using effective g factors, $\mu_{\text{sc}} = +1.59\mu_N$ is obtained, which agrees somewhat better with the experimental result. A core polarization calculation using the model of Noya, Arima, and Horie [25] yields $\mu_{\text{sp}}^{\text{eff}} = +1.68\,\mu_N$, again in good agreement with experiment. Finally, a recent calculation of the magnetic moment of ^{69}As in the interacting boson-fermion model yielded $\mu = +1.465\,\mu_N$ [4]. This deviates somewhat more from the experimental value but it should be noted that the authors of Ref. [4] were dealing with the properties of a rather extended set of nuclei in the $A \approx 70$ region and were not focusing on ^{69}As in particular.

An overview of the magnetic moments of the lowest $5/2^-$ state in the odd-A As isotopes is given in Table I. As can be seen, our result confirms, but with a better precision than in Ref. [7], that the proton configuration for ^{69}As is the same as for the heavier ^{71}As and ^{73}As. The fact that the magnetic moments for the lighter isotopes ^{69,71,73}As are a factor of about 2 larger than the ones for ^{75}As and ^{77}As, which agree well with the Schmidt value, is because of a rearrangement of the proton pairs when passing the magic neutron number 40 as was shown in Ref. [26] using the core polarization model of Ref. [27].

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