Quadrupole Interactions of the Short-lived β-Emitter ^{16}N in TiO_2

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Quadrupole interactions of ^{12,14}N in BN(hexagonal) crystal were studied by detecting β-NQR of ^{12}N and FT-NMR of ^{14}N, respectively. β-NMR of ^{16}N(I^π = 2$^-$, $T_{1/2}$ = 7.13 s) in MgO crystal was detected to determine the magnetic moment to be $|\mu(^{16}\text{N}: 2^-)| = (1.986 \pm 0.001)\ \mu_\text{N}$. Also, the β-NQR's of ^{12,16}N in TiO_2 crystal were detected to be $|Q(^{16}\text{N}: 2^-)| = (17.9 \pm 1.7)$ mb. An abnormally small effective charge for neutrons is required to account for $|Q(^{16}\text{N}: 2^-)|$.

Key words: Quadrupole Moments; N in TiO_2; FT- and β-NMR; Effective Charges of Nucleons in the Nucleus.

1. Introduction

Using recently developed β-NM(Q)R and isotope separators connected with particle accelerators, we have precisely measured the nuclear electromagnetic moments of many short-lived nuclei located around the proton and neutron drip-lines to obtain a deeper understanding of nuclear properties [1 - 3]. Those moments reveal new trends in nuclear shell structure, especially an extended radial distribution of the valence nucleons near the nuclear surface. For example, loosely bound valence nucleons to a core nucleus may affect the core less than the deeply bound ones.

Of particular interest is the nuclear structure of the ground state of ^{16}N(I^π = 2$^-$, $T_{1/2}$ = 7.13 s), which has a rather small one-neutron separation energy S_n = 2.49 MeV; the shell model predicts its simple structure as predominantly consisting of one neutron occupying the $d_{5/2}$-orbital outside the doubly closed ^{16}O core, and one proton-hole in the $p_{1/2}$-orbital inside the core. A definite understanding of the ground state can be inferred from the magnetic moment $\mu(^{16}\text{N}: 2^-)$ and quadrupole moment $Q(^{16}\text{N}: 2^-)$. In spite of such importance both, μ and Q are not known because of the experimental difficulties in maintaining the spin polarization of ^{16}N in suitable implantation media for long enough periods compared with its half life $T_{1/2}$ = 7.13 s. Another difficulty was that the two β-decay transitions from its ground state, i. e., $2^- \rightarrow 0^+$ and $2^- \rightarrow 3^-$, have similar decay rates with opposite signs of β-decay asymmetry factors A. Here we present the quadrupole interactions of ^{12,14}N in a BN crystal to determine $Q(^{12}\text{N}: 1^+)$ as the standard for short-lived N isotopes, and ^{12,16}N in a TiO_2 crystal which was used as a spin Dewar for the N isotopes to investigate their hyperfine interactions and finally to determine $Q(^{16}\text{N}: 2^-)$. Part of the present results has been reported in [4, 5].

2. Experiment

The present experimental method and setup used for the studies of ^{16}N were essentially similar to the previous β-NM(Q)R work on ^{12}N [1, 2, 4]. Polarized ^{16}N nuclei were produced through the ^{15}N(d, p)^{16}N reaction initiated with a deuteron beam of incident energy E_d = 2.5 MeV obtained from the van de Graaff at Osaka University. The Ti^{15}N target was prepared by nitriding a 0.5-mm thick titanium plate in enriched $^{15}N_2$ gas at T = 1250 K. The ^{16}N nuclei, leaving

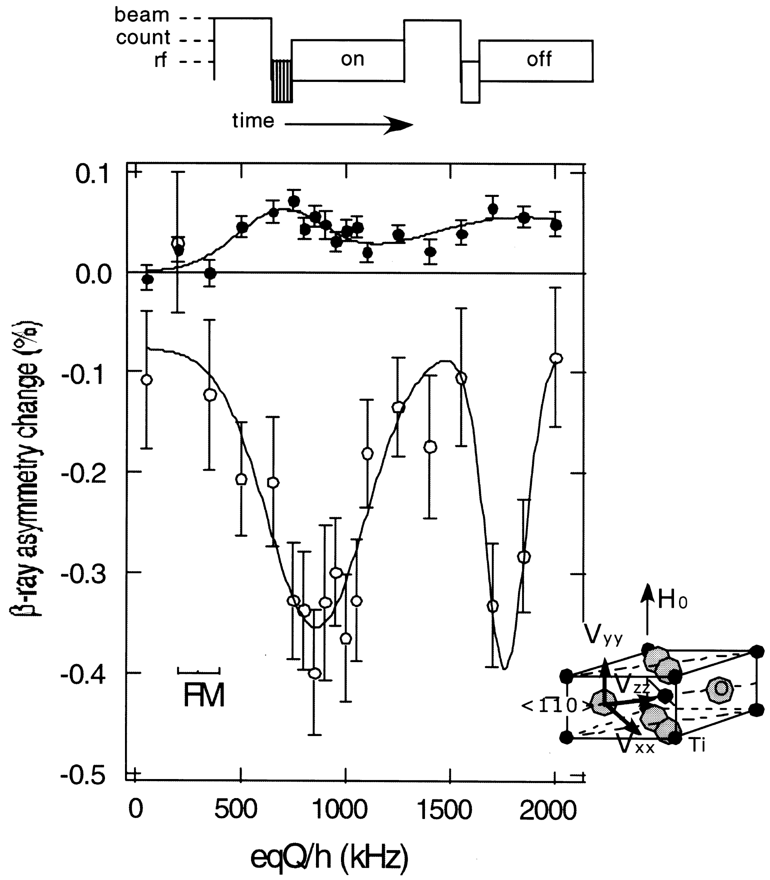

Fig. 1. Typical β-NQR spectra implanted in TiO_2 detected by the depolarization method. The c-axis of the crystal was set parallel to H_0, as shown in the inset. The full circles stand for the $2^- \rightarrow 3^-$ and the open circles for the $2^- \rightarrow 0^+$. The peak at the lower frequency is the normal β-NQR signal from ^{16}N at the substitutional oxygen site. The peak at the higher frequency is due to the double quantum transitions between the magnetic substate ± 2 and 0, these transitions were induced by the inner two preset rf's for an $eqQ/h \sim 1760$ kHz. The solid lines are the theoretical curves best fitted to the data.

Table 1. β-NQR detection of ^{16}N in TiO_2. Conditions are H_0 = 7.00 kOe, ν_L = 5302 kHz, $H_0 \sim$ 10 Oe, and frequency modulation width $FM = \pm$ 50 kHz.

	β Transition $2^- \rightarrow 0^+$ Single Q. T.	β Transition $2^- \rightarrow 0^+$ Double Q. T.[a]	β Transition $2^- \rightarrow 3^-$ Single Q. T.	β Transition $2^- \rightarrow 3^-$ Double Q. T.[a]
$\lvert eqQ(^{16}N \text{ in } TiO_2)/h\rvert$ (kHz)	858 ± 34	880 ± 14	683 ± 39	944 ± 120
Averaged (kHz)	877 ± 13		714 ± 37	
All Averaged	$\lvert eqQ(^{16}N \text{ in } TiO_2)/h\rvert$ = 859 ± 12(stat) |± 13(stat) (kHz)			
$\lvert eqQ(^{12}N \text{ in } TiO_2)/h\rvert$ [7, 5]	469 ± 5 (kHz)			
$\lvert Q(^{16}N; 2^-)\rvert$	17.9 ± 1.7 (mb)			

[a] Transition between magnetic substates $m = \pm 2 \rightarrow 0$.

the surface of the target on which the incident beam impinged over an angular range from 22 to 38° relative to the d-beam direction, were implanted into a catcher. The spin polarization of the implanted nuclei was about P_0 = 1.5% with a β-ray counting rate of $\sim 1 \times 10^4$/s measured with a set of β-ray counters and a deuteron beam intensity of 1 µA. To maintain the polarization of ^{16}N and to detect its β-NM(Q)R after implantation, a static magnetic field of H_0 = 7.0 kOe was applied parallel to the polarization.

To determine $\mu(^{16}N)$, the ^{16}N nuclei were implanted into an MgO crystal of $0.5 \times 20 \times 36$ mm^3, at room temperature. Also to determine $Q(^{16}N)$, the ^{16}N nuclei were similarly implanted into a TiO_2 single crystal of $0.5 \times 20 \times 36$ mm^3, the structure and the direction of the EFG of which are given in the inset of Figure 1. A new technique was also devised to separately detect the β-decay asymmetries in the two transitions, i. e., a set of detectors consisting of 3 plastic scintillators with one β-ray energy degrader inserted between the second and third scintillator. Each one was 1 mm thick with 30×30 mm^2 area. The degrader was a 5-mm thick aluminium plate with the same area. The integrated thickness of the first two scintillators and the degrader of a set was chosen such that all β-rays from the $2^- \to 3^-$ transition with E_β(max) = 4.0 MeV were stopped, but the β-rays with energies higher than 4 MeV from the $2^- \to 0^+$ transition with E_β(max) = 10.4 MeV could hit the third scintillator. The coincidence signals of the first and second scintillators gave 80% purity for the $2^- \to 3^-$ decay. The single counts from the third scintillator were ∼100% from the $2^- \to 0^+$ decay.

To measure μ, ^{16}N was produced during a beam-on production time of 8 s, followed by a beam-off time of 12.5 s for β-ray counting. At the end of each production time, an rf time of 20 ms duration was inserted before the next counting time started. The adiabatic fast passage technique was employed for inverting the spin polarization at on-resonance condition. Further-more, each counting time was divided into two with durations of 4.5 s and 8.0 s. Between the two we applied again an rf for 20 ms. A pair of beam-count cyclings were repeated to observe NMR, where one cycling started with rf and the other one with no rf. The FM width of the rf was 50 kHz. The 4-up/down-counting-rate ratios in the two beam-count cyclings gave $4A(P - P_0')$, where P is the residual polarization after applied rf. We found $|\mu(^{16}\mathrm{N}{:}\ 2^-)|$ = $(1.986 \pm 0.001)\mu_\mathrm{N}$.

To detect β-NQR of ^{16}N, a set of 4-rf fields ν_2, ν_1, ν_0, and ν_{-1} with a rf intensity H_1 = 10 Oe, a frequency modulation 50 kHz, and a duration period 2 ms, were applied in series. Here the ν_i's are the 4-rf transition frequencies at high field for a given ν_Q and η, which corresponds to site I defined in Section 4. The set was repeated 10 times in an rf time to depolarize the initial polarization at the on-resonance condition. The time sequence program is shown in the inset of Figure 1. A pair of beam-count cyclings, one with rf on and one without rf, were repeated until we obtained enough counting statistics. Thus the NQR effect Δ was $[(U/D)_\mathrm{on}/(U/D)_{off} - 1] \approx 2A(P - P_0'')$, where P_0'' is the polarization observed without rf for the ^{16}N in site I of TiO_2. In the typical spectrum given in Fig. 1, two rather broad peaks are shown, where the one at $eqQ/h \sim 860$ kHz corresponds to the single quantum transition of the substitutional ^{16}N, and the other at the higher $eqQ/h \sim 1750$ kHz corresponds to the double quantum transitions between $\pm 2 \to 0$ sublevels induced by the inner two $\pm 1 \to 0$ transitions present for $eqQ/h \sim 1720$ kHz. The preset ν_{-1}, and ν_1, hit the true transitions ν_2 (true) and ν_0 (true) simultaneously. The solid curve is the theoretical β-NQR line shape best fit to the data. The results are summarized in Table 1. We obtain a mean value as $|eqQ(^{16}\mathrm{N})/h|$ = $\{859 \pm 12(\mathrm{stat}) \pm 13(\mathrm{syst})\}$ kHz. The spread was $\delta eqQ/h$ = 150 kHz. Taking the symmetric nature of the line shape with the present precision, we added the systematic uncertainty of ±13(syst) kHz [5].

3. β-NQR of ^{12}N and FT-NMR of ^{14}N in BN Crystal

As the reference for short-lived N isotopes the quadrupole moment $Q(^{12}\mathrm{N})$ was determined by detecting β-NQR of $^{12}\mathrm{N}(I^\pi = 1^+, T_{1/2} = 11$ ms) implanted in a BN(hexagonal) crystal the c-axis of which was placed perpendicular to H_0 at room temperature [5]. As a result $|eqQ(^{12}\mathrm{N})/h|$ = (52.8 ± 4.1) kHz was obtained for the majority group (70%) ^{12}N implanted in N substitutional site in BN. In this measurement the parameters of the EFG, the directions of the principal components were measured beforehand by detecting the FT-NMR of ^{14}N in the crystal at 47 kOe at room temperature. The results have been obtained as $|eqQ(^{14}\mathrm{N})/h|$ = (110.7 ± 4.1) kHz, while η = 0 was assumed because of the crystal structure of BN, where the direction of q was parallel to the c-axis. Since $Q(^{14}\mathrm{N})$ = +(20.0 ± 0.2) mb had been known [6], the value q(N in BN) = $(2.29 \pm 0.09)\times 10^{20}$ V/m^2 was determined. Also the ratio $|Q(^{12}\mathrm{N})|/Q(^{14}\mathrm{N})$ = 0.477 ± 0.041 was obtained. Then the quadrupole moment of ^{12}N was determined as $|Q(^{12}\mathrm{N})|$ = (9.6 ± 0.8) mb.

4. Measurement of EFG in TiO_2

To obtain the EFG parameters of ^{12}N in TiO_2, NQR of ^{12}N in TiO_2 was detected for the two cases: For one case the crystal <001> was placed parallel to the direction of H_0, and for the other <1$\bar{1}$0> was placed parallel to H_0. To our surprise, the polarization as produced in the nuclear reaction was totally maintained (100%) in the TiO_2 crystal at external fields above 5.0 kOe at room temperature. Two independent final sites with almost equal ^{12}N populations were found. For one location (site I), a smaller EFG with eqQ/h = +(469 ± 5) kHz and η = 0.37 ± 0.02 was obtained. For the other location (site II), a larger EFG with eqQ/h = +(2888 ± 12) kHz and η = 0.038 ± 0.005 was obtained. In each site, the largest $|V_{ii}|$ of EFG was either q_a or q_b. In each set q_a (q_b) was parallel to either the <110> or <110> axis. The presently detected ^{12}N resided in diamagnetic circumstances. A possible final site is the substitutional site of an oxygen atom, where the ^{12}N atom is negatively charged, i. e., the neutrality was +1. One other possible site is the substitutional site of Ti where a ^{12}N is positively charged, i. e., the neutrality was –1. We can not reject the possibility that one site is an interstitial site in a unit cell with symmetry of the surroundings, where a ^{12}N atom was negatively or positively charged, i. e., the neutrality is +1 or -1.

5. Discussion

Using the known EFG and $Q(^{12}N)$ [5], we determined $|Q(^{16}N)|$ = (17.9 ± 1.7) mb as summarized in Table 1. The theoretical single particle value for the Q moment of the $|(\pi p_{1/2})^{-1}(\nu d_{5/2})^{+1}\rangle$ configuration, which occupies 96.1% of the total configuration gives $Q(^{16}N; jj)$ = –23 mb, where a harmonic oscillator potential (HO) with the oscillator length b = 1.76 fm and a standard effective charge, e_n^{eff} = +0.5e for neutrons in sd-shell nuclei [8], (Sagawa and Brown) are used. This is already ∼30% larger than the experiment. The shell-model-code OXBASH for p- and sd-model space gives Q_n(HO) = –47.5 e_n^{eff} mb and Q_p(HO) = –5.9 e_p^{eff} using the HO wave function for neutron and proton groups, respectively, i. e., without halo effect. Using the empirical effective charges e_n^{eff}(HO) = +0.48e for neutrons in sd-shell and e_p^{eff}(HO) = +1.48 e for protons in p-shell [9], the theory gives $Q(^{16}N$; HO) = –31.5 mb, which is 70% larger than the experimental value. This large discrepancy may indicate a crucial effect of the small binding energy on the neutron component. On the contrary, the Hartree Fock (HF) wave function with halo effect, gives Q_n(HF) = –60.4 e_n^{eff} and Q_p(HF) = –5.1 e_p^{eff}, to yield $Q(^{16}N$; HF+halo) = –27.3 mb by using empirical e_p^{eff}(HF) = +1.32 e and e_n^{eff}(HF) = +(0.34 ± 0.04)e [9]. The $Q(^{16}N$; HF+halo) is still 50% larger than the experiment. In order to reproduce the experimental value, a set of new effective charges must be introduced. Since $|Q_{exp}(^{16}N)|$ = 17.9 mb must be explained by the theoretical $|Q_n$(HF) + Q_p(HF)$|$ = $|$–60.4 e_n^{eff} + (–5.1)$e_p^{eff}|$, the e_n^{eff}(HF) values must be in the range of 0.19 e ∼ 0.21 e for which e_p^{eff}(HF) is within 1.3 e ∼ 1.0 e. Adopting the difference of the theoretical Q_n(HF)/e_n^{eff} and Q_n(HO)/e_n^{eff} as the theoretical uncertainty for the nuclear matrix, we conclude a definitely small effective charge for the neutron in the $d_{5/2}$ state of ^{16}N, e_n^{eff}(HF) = +(0.20 ± 0.04) e. The value is almost 40% of the systematic effective charges for a neutron in the sd-shell [8, 9]. The present small e_n^{eff}(HF) may indicate an important effect on effective charges for loosely bound neutrons, i. e., a relatively large $\langle r^2\rangle^{1/2}$ value of them and less perturbation to the core of the nucleus. Such a small neutron effective charge may also be found in ^{15}B [10], ^{17}B, ^{18}N [11] and ^{19}O [12], provided their nuclear structures will be well investigated.

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