Quadrupole Interactions of the Short-lived $\beta$-Emitter $^{16}$N in TiO$_2$

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Quadrupole interactions of $^{12,14}$N in BN(hexagonal) crystal were studied by detecting $\beta$-NQR of $^{12}$N and FT-NMR of $^{14}$N, respectively. $\beta$-NMR of $^{16}$N($I^T = 2^-, T_{1/2} = 7.13$ s) in MgO crystal was detected to determine the magnetic moment to be $\mu(16N: 2^-) = (1.986 \pm 0.001) \mu_N$. Also, the $\beta$-NQR's of $^{14}$N in TiO$_2$ crystal were detected to be $|Q^{(16N: 2^-)}| = (17.9 \pm 1.7)$ mb. An abnormally small effective charge for neutrons is required to account for $|Q^{(16N: 2^-)}|$. 

Key words: Quadrupole Moments; N in TiO$_2$; FT- and $\beta$-NMR; Effective Charges of Nucleons in the Nucleus.

1. Introduction

Using recently developed $\beta$-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($I^T = 2^-, T_{1/2} = 7.13$ s), which has a rather small one-neutron separation energy $S_0 = 2.49$ 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 $\mu(16N: 2^-)$ and quadrupole moment $Q(16N: 2^-)$. In spite of such importance both, $\mu$ 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 $\beta$-decay transitions from its ground state, i.e., $2^- \rightarrow 0^+$ and $2^- \rightarrow 3^-$, have similar decay rates with opposite signs of $\beta$-decay asymmetry factors $A$. Here we present the quadrupole interactions of $^{12,14}$N in a BN crystal to determine $Q^{(12N: 1^-)}$ 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 $Q^{(16N: 2^-)}$. 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 $\beta$-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...
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 $\beta$-ray counting rate of $\sim 1 \times 10^4/s$ measured with a set of $\beta$-ray counters and a deuteron beam intensity of 1 $\mu$A. To maintain the polarization of $^{16}$N and to detect its $\beta$-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 $\beta$-decay asymmetries in the two transitions, i.e., a set of detectors consisting of 3 plastic scintillators with one $\beta$-ray energy degrader inserted

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**Fig. 1.** Typical $\beta$-NQR spectra implanted in TiO$_2$ detected by the depolarization method. The $\epsilon$-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 $\beta$-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 $\pm 2$ and 0, these transitions were induced by the inner two preset rf’s for an $\epsilon \nu Q/\hbar \sim 1760$ kHz. The solid lines are the theoretical curves best fitted to the data.

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**Table 1.** $\beta$-NQR detection of $^{16}$N in TiO$_2$. Conditions are $H_0 = 7.00$ kOe, $\mu_0 = 5302$ kHz, $H_0 \sim 10$ Oe, and frequency modulation width $F/M = \pm 50$ kHz.

<table>
<thead>
<tr>
<th>$\epsilon Q(16N)/\hbar$ (kHz)</th>
<th>Single Q. T.</th>
<th>Double Q. T.</th>
<th>$\epsilon Q(16N)/\hbar$ (kHz)</th>
<th>Single Q. T.</th>
<th>Double Q. T.</th>
</tr>
</thead>
<tbody>
<tr>
<td>All Averaged</td>
<td>$858 \pm 34$</td>
<td>$880 \pm 14$</td>
<td>$683 \pm 39$</td>
<td>$714 \pm 37$</td>
<td>$944 \pm 120$</td>
</tr>
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</table>

$^a$ Transition between magnetic substates $m = \pm 2 \rightarrow 0$.
between the second and third scintillator. Each one was 1 mm thick with 30×30 mm² 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⁻→3⁻ transition with $E_β$ (max) = 4.0 MeV were stopped, but the β-rays with energies higher than 4 MeV from the 2⁻→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⁻→3⁻ decay. The single counts from the third scintillator were ∼100% from the 2⁻→0⁻ decay.

To measure μ, ¹⁶N 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 cycling 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 4Δ(P − P₀), where $P$ is the residual polarization after applied rf. We found $μ(¹⁶N; 2⁻→0⁻) = (1.986 ± 0.001)$%N.

To detect β-NQR of ¹⁶N, a set of 4-rf fields $ν₂$, $ν₁$, $ν₀$, and $ν₋₁$ with a rf intensity $H_r = 10$ Oe, a frequency modulation 50 kHz, and a duration period 2 ms, were applied in series. Here the $ν₁$'s are the 4-rf transition frequencies at high field for a given $ν₀$ and $ν₋₁$ 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 $|μ/D|_{on}|ν/D|_{off} = 1 ≈ 2A(P − P₀)$, where $P₀$ is the polarization observed without rf for the ¹⁶N in site I of TiO₂. In the typical spectrum given in Fig. 1, two rather broad peaks are shown, where the one at $eqQ/h ≈ 860$ kHz corresponds to the single quantum transition of the substitutional ¹⁶N, and the other at the higher $eqQ/h ≈ 1750$ kHz corresponds to the double quantum transitions between ±2 → 0 sublevels induced by the inner two ±1 → 0 transitions present for $eqQ/h ≈ 1720$ kHz. The preset $ν₋₂$ and $ν₋₁$, hit the true transitions $ν₋₂$ (true) and $ν₋₁$ (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 $μQ(¹⁶N)/h = \{859 ± 12(stat) ± 13(syst)\}$ kHz. The spread was $δμQ(¹⁶N)/h = 150$ 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 ¹²N and FT-NMR of ¹⁴N in BN Crystal

As the reference for short-lived N isotopes the quadrupole moment $Q(¹²N)$ was determined by detecting β-NQR of ¹²N($I^+=(1¹)$, $J=1/2$) ms) implanted in a BN(hexagonal) crystal the c-axis of which was placed perpendicular to $H₀$ at room temperature [5]. As a result $μQ(¹²N)/h = (52.8 ± 4.1)$ kHz was obtained for the majority group (70%) ¹²N 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 ¹⁴N in the crystal at 47 kOe at room temperature. The results have been obtained as $μQ(¹⁴N)/h = (110.7 ± 4.1)$ kHz, while $ν₋₁ = 0$ was assumed because of the crystal structure of BN, where the direction of $q$ was parallel to the c-axis. Since $Q(¹⁴N) = +(20.0 ± 0.2)$ mb had been known [6], the value $q(N in BN) = (2.29 ± 0.09) × 10^{20}$ V/m² was determined. Also the ratio $|Q(¹²N)|/|Q(¹⁴N)| = 0.477 ± 0.041$ was obtained. Then the quadrupole moment of ¹²N was determined as $|Q(¹²N)| = (9.6 ± 0.8)$ mb.

### 4. Measurement of EFG in TiO₂

To obtain the EFG parameters of ¹²N in TiO₂, NQR of ¹²N in TiO₂ was detected for the two cases: For one case the crystal <001> was placed parallel to the direction of $H₀$, and for the other <110> was placed parallel to $H₀$. To our surprise, the polarization as produced in the nuclear reaction was totally maintained (100%) in the TiO₂ crystal at external fields above 5.0 kOe at room temperature. Two independent final sites with almost equal ¹²N populations were found. For one location (site I), a smaller EFG with $eqQ/h = +(469 ± 5)$ kHz and $ν₋₁ = 0.37 ± 0.02$ was obtained. For the other location (site II), a larger EFG with $eqQ/h = +(2888 ± 12)$ kHz and $ν₋₁ = 0.038 ± 0.005$ was obtained. In
each site, the largest $|V_{E1}|$ of EFG was either $q_D$ or $q_B$. In each set $q_D$ ($q_B$) 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 $Q^{(12}\text{N})$ [5], we determined $|Q^{(16}\text{N})| = (17.9 \pm 1.7) \text{ mb}$ as summarized in Table 1. The theoretical single particle value for the $Q$ moment of the $\left|\langle t_2^{1/2}\|\nu_{d_{5/2}}\|^1\right|$ configuration, which occupies 96.1% of the total configuration gives $Q^{(16}\text{N}; j\bar{j}) = -23 \text{ mb}$, where harmonic oscillator potential (HO) with the oscillator length $b = 1.76 \text{ fm}$ and a standard effective charge, $\epsilon_n^{\text{eff}} = +0.5 e$ for neutrons in sd-shell nuclei [8], (Sagawa and Brown) are used. This is already $\sim 30\%$ larger than the experiment. The shell-model-code OXBASH for p- and sd-model space gives $Q_n^{(\text{HO})} = -47.5 \epsilon_n^{\text{eff}} \text{ mb}$ and $Q_p^{(\text{HO})} = -5.9 \epsilon_p^{\text{eff}}$ using the HO wave function for neutron and proton groups, respectively, i.e., without halo effect. Using the empirical effective charges $\epsilon_n^{\text{eff}}(\text{HO}) = +0.48 e$ for neutrons in sd-shell and $\epsilon_p^{\text{eff}}(\text{HO}) = +1.48 e$ for protons in p-shell [9], the theory gives $Q^{(16}\text{N}; \text{HO}) = -31.5 \text{ mb}$, 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 $Q_n^{(\text{HF})} = -60.4 \epsilon_n^{\text{eff}}$ and $Q_p^{(\text{HF})} = -5.1 \epsilon_p^{\text{eff}}$ to yield $Q^{(16}\text{N}; \text{HF+halo}) = -27.3 \text{ mb}$ by using empirical $\epsilon_n^{\text{eff}}(\text{HF}) = +1.32 e$ and $\epsilon_n^{\text{eff}}(\text{HF}) = +0.34 (\pm 0.04) e$ [9]. The $Q^{(16}\text{N}; \text{HF+halo})$ is still $50\%$ larger than the experiment. In order to reproduce the experimental value, a set of new effective charges must be introduced. Since $Q_{\text{exp}}^{(16}\text{N}) = 17.9 \text{ mb}$ must be explained by the theoretical $|Q_n^{(\text{HF})} + Q_p^{(\text{HF})}| = |60.4 \epsilon_n^{\text{eff}} + (-5.1) \epsilon_p^{\text{eff}}|$, the $\epsilon_n^{\text{eff}}(\text{HF})$ values must be in the range of $0.19 e \sim 0.21 e$ for which $\epsilon_p^{\text{eff}}(\text{HF})$ is within $1.3 e \sim 1.0 e$.

Adopting the difference of the theoretical $Q_n^{(\text{HF})}\epsilon_n^{\text{eff}}$ and $Q_p^{(\text{HO})}\epsilon_p^{\text{eff}}$ 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 $^{16}\text{N}$, $\epsilon_n^{\text{eff}}(\text{HF}) = +(0.20 (\pm 0.04) e$. The value is almost $40\%$ of the systematic effective charges for a neutron in the sd-shell [8, 9]. The present small $\epsilon_n^{\text{eff}}(\text{HF})$ 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 $^{15}\text{B}$ [10], $^{17}\text{B}$, $^{18}\text{N}$ [11] and $^{19}\text{O}$ [12], provided their nuclear structures will be well investigated.

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