

Electric Quadrupole Interactions of the Short-Lived β -Emitter ^{12}N in Insulator Crystals (^{12}N Implanted in Single Crystal TiO_2)*

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The electronic structure of nitrogen atoms as impurities in an ionic TiO_2 crystal has been investigated by analyzing electric field gradients (EFGs) measured by use of short-lived β -emitting ^{12}N implanted following nuclear reactions. Conventional β -NMR and its modification, suitable for the detection of quadrupole effects in the NMR spectra, were used for the investigation of hyperfine interactions of ^{12}N located in substitutional sites of O atoms and interstitial sites in the crystal. In order to deduce absolute values of the EFGs from the obtained eqQ/h , the quadrupole moment of ^{12}N has been determined from the NMR detection of ^{12}N implanted in BN(hexagonal) crystal. Here the EFG at the N atom in BN was measured by detecting the FT-NMR of ^{14}N in the crystal. The EFGs in TiO_2 are compared with the theoretical predictions based on the *ab initio* band-structure calculation in the framework of the KKR method.

Introduction

The electric quadrupole interaction of nuclides with crystalline internal fields discloses the spatial distribution of electronic and nucleonic charges. Of particular interest are the electronic structures of very dilute impurities in the substitutional and interstitial sites of the crystalline unit cells. Such studies have become practical owing to the recent development of NMR techniques applicable to short-lived nuclides, which enables to get very microscopic information on solids, i.e., especially, the dynamics associated with the formation of probe-host complexes, and that of nuclides that can not be investigated by use of the natural isotopes due to the limitations in their natural abundances and nuclear moments [1–3]. Investigations on the dynamics may guide us to discover fundamental interactions and/or a new coexistence of the impurities with the modified host ambient [2, 4]. In addition to such studies, the acquired knowl-

edge may help us in synthesizing new and strange materials that are longed for in industrial applications.

Besides such investigations in the material sciences, a surprising application of the technique, in which the well defined quadrupole interactions of the nuclides in single crystals are used, arises in particle and/or nuclear physics; it provides us with a tool for manipulating spin orientations of the particles and nuclides as well as with a treasure box, a unit cell in crystals, as a container of the short-lived polarized nuclides. As a result, enables us to perform correlation-type measurements for extracting new fundamental information that has been impossible to be squeezed out of other methods, i.e., the correlation between the spin orientations and the out going nuclear radiations from short lived nuclides implanted in the crystals, that are forced to be exposed artificially to the internal field superimposed to the externally applied magnetic field [5]. Recent findings from such high precision measurements of the nuclear moments and β -decays have been non-nucleonic degrees of freedoms in the nucleus, e.g., quantitative studies of mesonic and even quark effects in nuclei.

In the present article we introduce two experimental techniques, the conventional β -NMR technique following low-energy nuclear reactions [6], and the modified one specifically suitable for the detection of quadrupole interactions of nuclides with high nuclear spins [7]. In the next step, we discuss the recent developments in creating spin polarized nuclides through high-energy

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heavy-ion reactions, the implantation process following nuclear reactions [8], the hyperfine interactions of short-lived β -emitting ^{12}N nuclei in an ionic crystal TiO_2 [9], and finally a comparison of the measured electric field gradients (EFGs) with the theoretical predictions based on the *ab initio* band-structure calculation in the framework of the KKR method in which the local lattice relaxation is taken into account [1].

Experimental Method

1. Conventional β -NMR

The experimental set up employed in the present study is schematically shown in Figure 1. In the figure, in order to grasp the basic idea of the method easily, the magnet to employ the high field H , the vacuum chamber in which the whole system is included, the target holder, and the target chamber to which a recoil collimator is mounted are not shown. The conventional β -NMR method consists of the following five steps [6]: 1) the production of spin polarized short-lived β emitters through nuclear reactions, 2) the implantation of the nuclides into crystals using the kinetic energy obtained in nuclear reactions, 3) the preservation of the polarization in the crystals during the lifetime of the nuclides, 4) the detection of the β -decay asymmetry by detecting the β -ray angular distribution, 5) the manipulation of the polarization with rf magnetic fields (NMR) and the following detection of the asymmetry changes in the β -ray distribution.

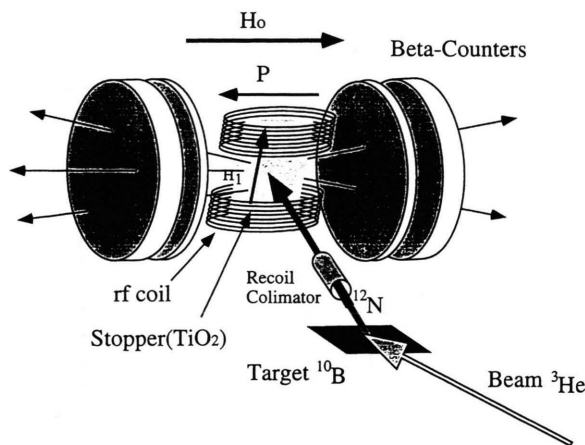


Fig. 1. Schematic view of the conventional β -NMR device. In order to see the method clearly, the magnet to employ the high field H , the vacuum chamber in which the whole system is included, the target holder, and the target chamber to which recoil collimator is mounted are not shown.

The ^{12}N ($I^\pi = 1^+$, $T_{1/2} = 11$ ms) nuclei are produced through the $^{10}\text{B}(^3\text{He}, n)^{12}\text{N}$ reaction with a ^3He beam of 3.0 MeV obtained from the Van de Graaff accelerator at Osaka University. Selecting the ^{12}N nuclei ejected at 12° to 28° , a spin polarization of $P \sim 0.25$ was obtained, which was maintained in a disk of TiO_2 single crystal the size of which was $0.3 \times 20 \times 20 \text{ mm}^3$. The polarized ^{12}N nuclei were implanted by use of their recoil energy given through the reaction into the crystal, with its crystal c -axis placed parallel to a strong external magnetic field of $H = 7.3 \text{ kOe}$ which was also parallel to the polarization. As shown here, in this conventional method, nuclear reactions initiated with light incident particles of less than 10 MeV are usually used to produce β emitters, i.e., no isotope separators are needed in many cases because of the relatively small amount of background activities with similar or longer lifetimes of ^{12}N produced in the reaction [6]. This cool condition was reinforced by employing a pulsed beam method, i.e., the β rays were counted during only the counting time without the beam on the target as shown in the insert of Figure 3. By the way, this recoil selection technique has been drastically expanded in its ability of producing various polarized β -emitters near the proton and neutron drip lines by employing the projectile fragmentation process in high-energy heavy-ion reactions initiated with an incident energy of up to 250 MeV/nucleon as the fragment separator developed [8] at Lawrence Berkeley Laboratory (LBL) in U.S.A. is shown in Figure 2.

The angular distribution of β rays emitted from polarized ^{12}N as a function of the electron ejection angle θ defined relative to the direction of the polarization P is given as $W(\theta) = \{1 + P \cos \theta\}$. Here, a polarization is defined as $P = (\sum m p_m) / I(\sum p_m)$, where m is the magnetic substate and p_m is its population. Since we manipulate the spin polarization by the β -NMR technique to change the polarization, we detect the succeeding change in the asymmetric β -ray distribution [6]. In order to measure the β -decay asymmetry, β rays from ^{12}N implanted in the catcher were detected by two sets of plastic-scintillator-counter telescopes placed along the polarization direction such that one is above and the other below the catcher. The β rays, as mentioned already, were detected only during beam-off counting times, typically a duration of 40 ms, following a beam-on production time of 15 ms. This pulsed-beam method, the timing program of which is shown in Fig. 3, was to detect β -rays from ^{12}N during the counting time with the beam-off, i.e., after short-lived (shorter than ^{12}N) background activities almost died away, and to make the spin manipulation

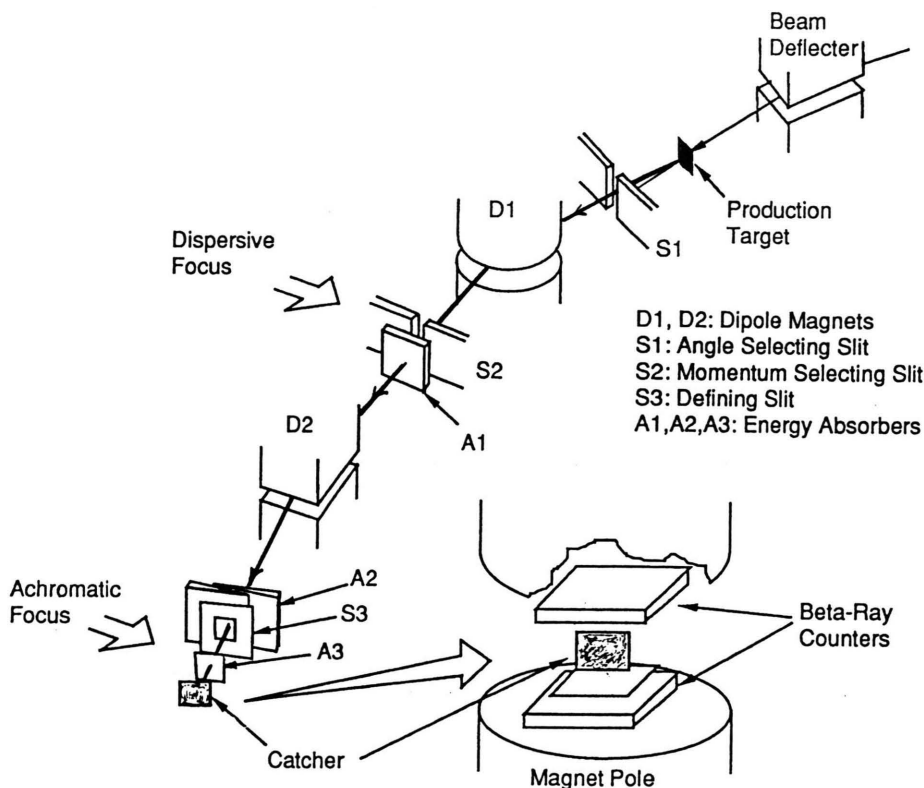


Fig. 2. Fragment separator following high energy heavy-ion reactions mated with the β -NMR equipment [8]. The incident energy of the heavy-ion was below 250 MeV/nucleon.

possible. The rf for the spin manipulation (NMR) was applied for 5 ms in between a production time and the following counting time. The up-down counting rate ratio of the time section for which the polarization was manipulated by the rf (rf-on) was normalized by the other ratio for the section without any manipulation (rf-off) as $\Delta R = \{W(0)/W(\pi)\}_{\text{on}} / \{W(0)/W(\pi)\}_{\text{off}} - 1 \sim 2(P_{\text{on}} - P_{\text{off}})$, where, P_i are the time averaged polarizations.

2. Quadrupole Interaction in High-Field

In electromagnetic fields, the energy E_m of a magnetic substate m is given by the sum of a magnetic coupling μH and an electric one, eqQ . In the high magnetic field limit, the energy is given to first order by perturbation theory [10] as

$$E_m = -(m/Ih)\mu H + (h/4)v_Q \cdot (3\cos^2\beta - 1 + \eta \cdot \sin^2\beta \cdot \cos^2\alpha)(m^2 - a/3). \quad (1)$$

Here $-\mu H$ is the magnetic interaction of the magnetic moment μ with a magnetic field H . The quadrupole

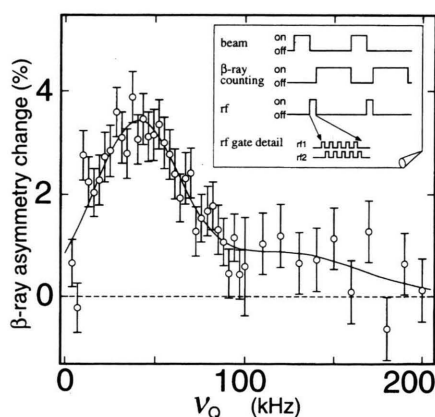


Fig. 3. Typical quadrupole spectrum of ^{12}N in BN crystal. The NMR of ^{12}N was detected by use of the modified β -NMR (β -NQR). The parameters of the field gradient were determined from the FT-NMR detection of ^{14}N in BN. The frequency of each rf was modulated ± 2 kHz. The solid line is the theoretical curve composed of two Gaussian components best fit to the data. The insert is the timing program of the pulsed beam method for the β -NMR.

coupling constant ν_Q and other parameters are defined as

$$\begin{aligned} \nu_Q &= \{3eqQ/2I(2I-1)h\}, \quad q = V_{zz}, \\ \eta &= (V_{xx} - V_{yy})/q, \quad \text{with } |V_{zz}| \geq |V_{yy}| \geq |V_{xx}|, \\ Q &= \text{nuclear quadrupole moment}, \\ I &= \text{nuclear spin } (=1 \text{ for } ^{12}\text{N}), \quad \text{and} \\ a &= I(I+1). \end{aligned} \quad (2)$$

Here, β and α are Euler angles of the EFG relative to the direction of H . In order to extract the quadrupole and magnetic coupling constants by use of the conventional β -NMR, a set of 2 rf transitions for the present spin $I=1$ among magnetic substates $m=\pm 1$ and 0 need to be separately measured (NMR spectrum). Such measurements are relatively difficult because of the very small β -NMR effects given by a single rf application compared with the NMR effect for the case without any quadrupole interaction.

In order to enhance the observable NMR effects in a β -NMR spectrum, the conventional β -NMR described above has been modified as a new nuclear quadrupole resonance (β -NQR) [7]. With this β -NQR detection used in the eqQ/h measurement of ^{12}N in BN, during each rf-on time the two transition frequencies for a given eqQ/h were applied in series, and the pair was repeated typically 5 times within an rf-on time. A typical time duration for a transition frequency was 1.0 ms, and that for the pair was 2.05 ms in which 25 μs was used for a change of frequency. Each rf was linearly modulated once in frequency with a range of ± 10 kHz. With a choice of a proper set of rf intensities, the polarization can be destroyed completely at the set of frequencies for the real eqQ/h . Suppose ten values of ν_Q were mapped in a ν_Q -spectrum measurement, and three rf-off cycles were added for normalization, (10+3) complete beam-count cycles were repeated until sufficient counting statistics were attained. The rf cycling system as well as the beam-count cycling system were controlled by a micro computer. The data taking system was also synchronized with the beam and rf cycling using the same computer.

3. Implantation of ^{12}N in TiO_2

Significant features in the implantation are described here. The depth of the implanted nuclei (the impurities) in the crystal is from its surface to about several μm for the use of nuclear reactions initiated with light ions of energies lower than ~ 10 MeV. The density of the impurities after their stop is very dilute, i.e., less than $10^{12}/\text{cm}^3$ which implies that any interactions among

the implanted impurities are negligible. Finally, the time range in which we can detect the interactions is governed by the nuclear lifetime of ^{12}N , i.e., 11 ms and its spin-lattice-relaxation times in the materials we use. For the applications of other short-lived nuclides in solids, the longer side is limited by their spin-lattice-relaxation time in catchers, which can be as long as ~ 100 s.

It has also been shown that the final sites of the implanted light nuclides are substitutional and interstitial sites in a crystalline unit cell. Also, the close neighbourhood of a final site, within the sphere determined by a radius of almost several 10 s lattices, is almost free at low temperature from any sea of damages and distortions produced by the traveling ions and other accompanied radiations, i.e., this has been observed at and below room temperature for almost all combinations of any nuclides and crystals we have studied [6, 11]. This is easily understood from the energy-loss process of traveling ions in matter; the rapid decrease, down to almost zero, of the energy loss of traveling ions in crystals at energies below 10 keV/nucleon (for ions with atomic number Z less than 20). Even at the lower energy, however, where ions do not lose their kinetic energy any more through ionization of the host atoms, still the ions keep moving forward without producing any damages along their paths in the crystal while they continue to lose their energy through the interaction with the phonons in the material. Finally the ions find their final sites. According to the systematic studies on the final sites for ions lighter than Sc ions in ionic crystals, a nuclide tends to substitute mainly with an atom which has the same Z number as the implanted ions, that are one of the constituents in the ionic crystal. Besides this, a small fraction, less than a half of the implanted ions, will find an interstitial site in the crystalline unit cell where the polarization of the implanted ions could not survive because of the paramagnetic atomic state. This happened in tests we have studied until now. The rate of this substitution depends on the atomic density of the isotope originally included in the crystal; i.e., the survival rate of the implanted nuclides in such ionic crystals is usually less than 50% even for favorable cases. In general, the spin-lattice-relaxation times of the implanted ions with diamagnetic surroundings in ionic crystals have been known to be fairly long, as long as $T_1 = 10 \sim 100$ s, which depends on each combination of the ions and crystals. It has also been discovered that a nuclide can substitute with an atom which has the atomic number $Z \pm 1$, that is one of the constituents in an ionic crystal [3]. On the other hand, the implanted ions lighter

Table I. Experimental and theoretical field gradients in TiO_2 . Among the principal components the value q_c is parallel to the crystalline c -axis, and q_a or q_b is parallel to the $\langle 1, 1, 0 \rangle$ or $\langle 1, \bar{1}, 0 \rangle$ axes, respectively. The field gradients were given for the singly ionized $\text{N}^\pm$ states in each final site. Sub. and Int. stand for substitutional and Interstitial sites, respectively.

Experimental results in units of 10^{19} V/m^2			
	q_a or q_b		q_c
Site I	$\mp 62 \pm 4$	$\pm 198 \pm 14$	$\mp 135 \pm 10$
Site II	$\mp 586 \pm 73$	$\pm 1220 \pm 85$	$\mp 633 \pm 44$
Theoretical results in units of 10^{19} V/m^2			
Site	q_a	q_b	q_c
Sub. $\text{Ti}(\text{N}^+)$	-46	-192	+239
Sub. $\text{O}(\text{N}^-)$	+185	-178	-6
Int. $(\text{N}^+)^*$	-449	+1304	-855
Int. $(\text{N}^-)^*$	-696	+2124	-1429

* Octahedral interstitial site; a face center of a unit cell.

than Ti in metals that we have studied similarly find their final sites mainly in the substitutional sites irrespective of the difference of the atomic number Z , and the rest of them 1 finds in interstitial sites [6, 11]. In these metal cases, the ions in both sites keep their polarizations, and their spin-lattice-relaxation times are usually so short that they are mainly governed by the Knight-shift interaction of the ions with conduction electrons of s -symmetry in metals.

For the studies on ^{12}N , it has been exceptionally difficult so far to find out suitable ionic crystals which contain N isotopes, in which ^{12}N ions have long spin-lattice-relaxation times and sharp NMR-line widths [12]. One of the reasons for these unsuccessful cases might be that in the nitrogen compounds the close surroundings of the final sites are not free from the implantation induced radiation damages and from the inherent nitrogen defects.

Measurement of the Quadrupole Moment of ^{12}N ($I = 1$, $T_{1/2} = 11 \text{ ms}$)

For the first step of the present studies, the quadrupole moment of ^{12}N was determined by detecting the β -NQR spectrum of ^{12}N implanted in a BN(hexagonal) crystal, the c -axis of which was placed perpendicular to H_0 at room temperature. In this run, the parameters of EFG, the directions of the principal components and the amount of the asymmetry η were measured by detecting the FT-NMR of ^{14}N in the crystal. Almost half of the im-

planted ^{12}N maintained their polarization in BN. A typical β -NQR spectrum is shown in Fig. 3, where the solid curve is the theoretical line shape which consists of two theoretical β -NQR shapes best fit to the data, where symmetric EFGs were assumed by taking the results obtained in a separate FT-NMR run of ^{14}N in BN, while spreads in the EFGs $\delta q/q \sim 60\%$ and $\sim 30\%$ for the majority and minority groups, respectively, were also obtained. The line spreads are theoretically analysed elsewhere [13]. As a result, $|eqQ(^{12}\text{N})/h| = 54.0 \pm 3.2 \text{ kHz}$ was obtained for the majority group (70%) ^{12}N in BN. For the minority group (30%), $|eqQ(^{12}\text{N})/h| = 175 \pm 19 \text{ kHz}$. From the systematic studies stated above, we conclude that the majority-group ^{12}N resides in the substitutional site of nitrogen atoms in BN. The residual minority group may reside in the substitutional site of B in BN with charge state N^{2+} , which has to be confirmed still.

Before the ^{12}N run and the theoretical prediction of line shapes of the β -NQR, the parameters of the EFG effective for the N atom in the BN crystal was measured by detecting FT-NMR of ^{14}N at 4.7 T and at room temperature. The results have been obtained as $|eqQ(^{14}\text{N})/h| = 110.7 \pm 4.1 \text{ kHz}$, while $\eta = 0$ was assumed because of the crystal structure of BN, where the direction of q was parallel to the c -axis. Since the quadrupole moment of the ^{14}N has been known as $Q(^{14}\text{N}) = +20.0 \pm 0.2 \text{ mb}$ [14], the value $q(\text{N in BN}) = (2.29 \pm 0.09) \times 10^{20} \text{ V/m}^2$ was determined.

What we need is the ratio of the two quadrupole moments of the two isotopes, $|Q(^{12}\text{N})|/Q(^{14}\text{N}) = 0.477 \pm 0.041$. Then the quadrupole moment of ^{12}N has been deduced using the known $Q(^{14}\text{N})$ values as $|Q(^{12}\text{N})| = 9.8 \pm 0.9 \text{ mb}$ [13].

NMR Detection of ^{12}N in TiO_2 Crystal

To obtain a complete knowledge on EFG parameters, the NMR spectra of ^{12}N were obtained for two cases: For one case the crystal c -axis $\langle 0, 0, 1 \rangle$ was placed parallel to the direction of H_0 , and for the other case the $\langle 1, 1, 0 \rangle$ axis was placed parallel to the direction of H_0 . First of all, to our surprise, the polarization as produced in the nuclear reaction was totally maintained (100%) in the TiO_2 crystal at external fields above $H = 0.5 \text{ T}$ and at room temperature. This never happened in other cases in which ionic crystals were used. Another surprise was that we found two independent final sites with almost equal ^{12}N populations, for which two sets of quadrupole splits

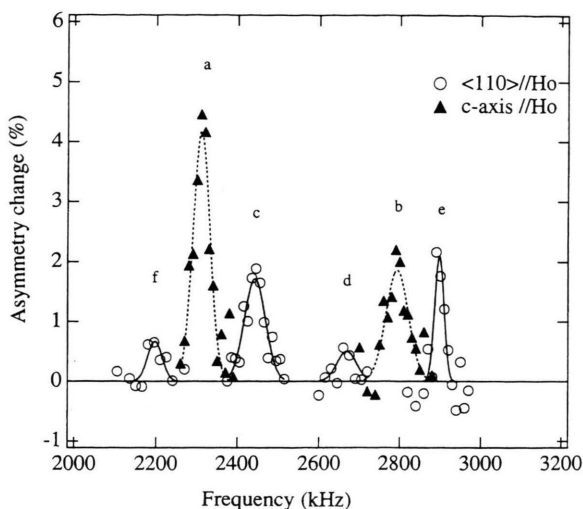


Fig. 4. Quadrupole spectra of ^{12}N in Site I in TiO_2 . Closed triangles: The c -axis was parallel to the external field $H=0.5$ T. One pair of resonances a and b with narrow split was observed. Open circle: The $\langle 1, 1, 0 \rangle$ axis was parallel to the external field $H=0.5$ T. The solid lines are Gaussian curves best fit to the data. Two pairs of resonances c and d, and e and f were observed.

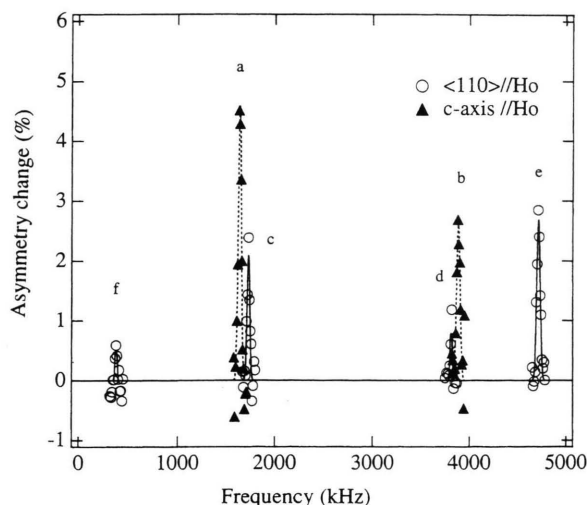


Fig. 5. Quadrupole spectra of ^{12}N in Site II in TiO_2 . Closed triangles: The c -axis was parallel to the external field $H=0.5$ T. One pair of resonances a and b with wide split was observed. Open circles: The $\langle 1, 1, 0 \rangle$ axis was parallel to the external field $H=0.5$ T. The solid lines are Gaussian curves best fit to the data. Two pairs of resonances c and d, and e and f were observed.

with narrow line widths were observed, i.e., two locations with equal ^{12}N populations were discovered. Typical NMR spectra for each ^{12}N group are shown in Figs. 4 and 5. For one location (site I), a relatively small EFG

Table II. Polarizations of short-lived β -emitters maintained in ionic crystals. The ratio of the maintained polarization P in crystals observed at room temperature divided by the initial one P_0 of the ions before implantation are given in %. a) Nuclear Physics **A559**, 239 (1993). b) Phys. Lett. **61B**, 155 (1976). c) Phys. Rev. Lett. **34**, 1465 (1975). d) Nucl. Phys. **A236**, 416 (1974). e) Phys. Rev. Letters **69**, 2058 (1992). f) To be published in Phys. Lett. g) Hyperfine Interactions **78**, 117 (1993). h) OULNS (Ann. Rep. Osaka Univ. Lab. Nucl. Studies) 1995, 69 (1996). i) Nuclear Physics **A559**, 239 (1993). j) The present results; OULNS (Ann. Rep. Osaka Univ. Lab. Nucl. Studies) 1995, 62 (1996).

Nucl.	I	$T_{1/2}$	Catcher	P/P_0	Ref.
^{41}Sc	7/2	0.54 s	CaO	50%	a
^{39}Ca	3/2	0.6 s	KBr, KCaBr ₃	~50	b
^8Li	2	0.84 s	LiIO ₃	30	c
^{17}F	5/2	66 s	CaF ₂ , NaF, MgF ₂	~50	d
^{12}B	1	21 ms	BN	30	e
^{12}N	1	11 ms	BN	30	f
^{23}Mg	3/2	11 s	TiO ₂ , MgF ₂	30	g
^{19}O	5/2	26.9 s	TiO ₂	30	h
^{41}Sc	7/2	0.54 s	TiO ₂	60	i
^{12}N	1	11 ms	TiO ₂	100	j

with a coupling constant $eqQ/h = +469 \pm 5$ kHz and $\eta = 0.37 \pm 0.02$ was obtained. For the other location (site II), on the contrary, a larger and near symmetric EFG with $eqQ/h = +2888 \pm 12$ kHz and small $\eta = 0.038 \pm 0.005$ was obtained. The plus signs for the two coupling constants were determined from the knowledge on the directions of the polarization, the applied H , and the plus sign of the spin alignment of the ^{12}N nuclei produced in the nuclear reaction. Using the present $|Q(^{12}\text{N})| = 9.6 \pm 0.8$ mb, we had two sets of EFGs and their parameters. The results are given in Table II. In each site, the largest component among the absolute values of the principal components of the EFG was either q_a or q_b . It was found that the component q_a (q_b) in each parameter set was parallel to either $\langle 1, 1, 0 \rangle$ or $\langle 1, \bar{1}, 0 \rangle$ axis.

Since each frequency that comes from the magnetic interaction only for each location is at the Larmor frequency, i.e., they resided in diamagnetic circumstances, a possible final site for a ^{12}N atom is reasonably expected to be the substitutional site of an Oxygen atom, where the ^{12}N atom is negatively ionized and as if it were an oxygen atom, i.e., the neutrality was +1. One other possible site is the substitutional site of Ti where a ^{12}N atom was positively ionized and pretended to be a Ti atom, i.e., the neutrality was -1. We can not reject the idea that one site is the interstitial site in a unit cell with the symmetry of the surroundings that reproduce the field gradient parameters here obtained, where a ^{12}N atom was negatively or positively ionized, i.e., the neutrality is +1 or -1.

ab initio Band Structure Calculation

The EFGs obtained here are compared with theoretical predictions based on the *ab initio* band-structure calculation in the framework of the KKR method [1]. To calculate the EFGs at the impurity sites, the super cell method was adopted. Here, a super cell consisted of two unit cells of the TiO_2 crystal, stacked along the c -axis with the body centered Ti atom, or one of the oxygen atoms is substituted with the impurity N atom. Regarding a possible interstitial site, the center of a face of a unit cell was chosen for the present trial considering the symmetry of the surrounding distribution of atoms. For the first step, in order to predict the lattice renormalization, the total energy of the super-lattice was evaluated as a function of the lattice relaxation. In this minimization of the energy, the direction of the shift of an atom was on the vector formed by the impurity and an atom which is going to be relaxed. For Ti or the interstitial site, the nearest 6 oxygen atoms that form the octahedron, and for the oxygen site the nearest three Ti atoms were taken into account. The rates of the relaxations $\delta a/a = -8\%$, $+0.3\%$ and 22% were predicted for N impurities at a substitutional Ti site, oxygen site, and octahedral interstitial site, respectively. In the next step, to interpret the diamagnetic circumstances of the ^{12}N in TiO_2 crystal, an effective charge compensation mechanism for the difference of the number of the valence electrons between oxygen and nitrogen was considered. Using the results on the charge density in TiO_2 obtained by the *ab initio* band structure calculations, EFGs at each ^{12}N site were calculated as given in Table II together with the experimental data. The field gradients for each site were evaluated as functions of the neutrality of the impurity N.

Discussion

The experimental site I was proposed to be the substitutional site of O atom in TiO_2 crystal, where the nitrogen atom is singly and negatively ionized N^- because the

theory on it reproduced the experimental result that q_a or q_b that lies in the $(0, 0, 1)$ plane has the largest absolute value among the principal components, as well as the relative signs of q_a and q_b are opposite. The theory also explains that q_c is parallel to the c -axis. Also the experimental site II was proposed to be the octahedral interstitial site of a unit cell where the nitrogen atom is singly and positively ionized N^+ because the theory on it reproduces the experimental result that one of those q_a or q_b that lies in the $(0, 0, 1)$ plane, has the largest absolute value among the principal components, as well as their relative signs are opposite. Even more, the theory explains the experimental largest absolute value. In all cases, however, the agreement of the other experimental components with the predictions was still poor. One of the reasons of these quantitative discrepancies may be in the huge difference between the real impurity density (10^{12} impurities/ cm^3) and that of the imaginary matter composed of superlattices ($\sim 10^{21}$ impurities/ cm^3).

Until the present implantation experiment on ^{12}N has been carried out, no short-lived nuclides have been known that totally maintained their spin polarizations in ionic crystals following nuclear reactions. In Table II, typical survival rates of such known cases are listed together with the present ^{12}N to see the systematic trends. The present combination of N- TiO_2 is clearly an exceptionally favorable case. Even if this success in maintaining the total polarization occurred by chance, the mechanisms as such are unique and interesting, and have to be clarified.

Further theoretical progress is encouraged to explain the above phenomena especially in the selections of Maf-fin-Tin potentials, the derivation of the lattice relaxations, the choice of super-lattices, and finding the interactions among implanted impurities and host atoms.

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