Development of a Nuclear Spin Dewar: Hyperfine Interactions of the Short-Lived β Emitter ^{12}B in TiO_2

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The β-NMR detection of ^{12}B ($I^\pi = 1^+$, $T_{1/2} = 20.2$ msec) implanted in a TiO_2 (rutile) single crystal following a nuclear reaction showed that its spin polarization produced in the reaction is totally maintained during its lifetime. Two implantation sites with relative populations 9:1 were identified. The electric field gradients (EFGs) were determined to be $q = +(37.1 \pm 0.5)\ 10^{15}$ V/cm^2 with $\eta < 0.03$ and $q = +(185 \pm 5)\ 10^{15}$ V/cm^2 with $\eta = 0.62 \pm 0.02$ for the major (90%) and minor (10%) sites, respectively. The EFGs were compared with the theoretical values given by the band-structure calculation in the framework of the KKR method. TiO_2 crystals with proper treatment can be a good "Spin Dewar" in which any short-lived nuclei can be implanted, and their spin polarizations as produced in nuclear reactions can be maintained during their lifetime.

Key words: Nuclear Polarization; β-NMR; Electric Field Gradient; TiO_2.

1. Introduction

The investigation of hyperfine interactions of short-lived nuclei implanted in crystal lattices provides new information on electronic structures of impurities and nuclear moments. Preservation of nuclear polarization is important in this regard, for the effective use of the β-NM(Q)R method which utilizes the asymmetric β-ray angular distribution from the spin polarized nuclear probes. We have shown that some of the ionic crystals maintain the polarization of β-emitting nuclei implanted in them [1]. These crystals are useful for a variety of experiments such as high precision measurements of nuclear moments and β-ray angular distributions which can probe non-nucleonic degrees of freedom inside the nucleus. Hence crystals which maintain the polarization produced in nuclear reactions of any implanted nuclei are of considerable interest. Requirements for such a "Spin Dewar" are as follows: 1.) preservation of the polarization without loss of any implanted nuclei produced in the reaction, 2.) longer spin-lattice relaxation times than their nuclear lifetimes, and 3.) a well defined small number of final implantation sites. The third requirement is important for the efficient manipulation of nuclear spin ensembles.

Recently we have found that TiO_2 (rutile) single crystal is one of the best implantation media as a spin Dewar, since many short-lived β emitters, such as ^{12}N [1], ^{13,19}O [2], and ^{41}Sc [3] maintained their full polarization produced through nuclear reactions after being implanted in the crystal at room temperature for as long as 10 sec or even longer. In the present study, the hyperfine interactions of the β-emitting nucleus ^{12}B ($I^\pi = 1^+$, $T_{1/2} = 20.2$ ms) implanted in TiO_2 are observed by use of the β-NM(Q)R method towards a systematic study to develop the Spin Dewer.

2. β-NMR Measurement

The experimental setup and procedure are similar to previous ones [1]. ^{12}B nuclei were produced

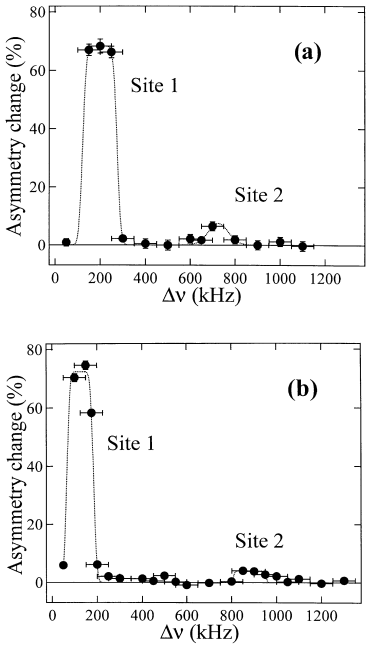

Fig. 1. Typical β-NQR spectra of ^{12}B in TiO_2 observed at 300 K for (a) H_0 = 5 kOe // c-axis, (b) H_0 = 5 kOe // $\langle 110 \rangle$ axis. RF magnetic field H_1 = 20 Oe with 100 kHz modulation width.

Table 1. EFGs st two different sites for ^{12}B in TiO_2.

	Site 1	Site 2
Population	90%	10%
$q \cdot 10^{15}$ V/cm^2	$+37.1 \pm 0.5$	$+185 \pm 5$
η	$< 0.03^{\#}$	0.62 ± 0.02

The upper limit was estimated from the frequency modulation width of rf.

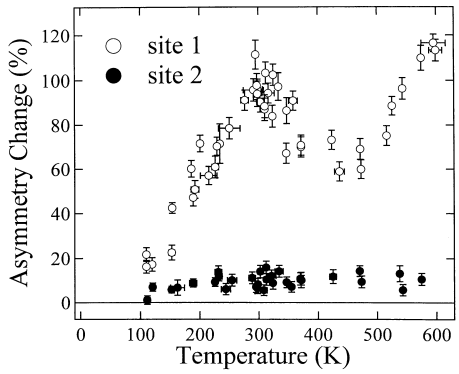

Fig. 2. Temperature dependence of the asymmetry change for ^{12}B in TiO_2. Open circles correspond to site 1 ($\Delta\nu$ = 200 $\pm$ 50 kHz) and closed circles to site 2 ($\Delta\nu$ = 750 $\pm$ 50 kHz).

through nuclear reaction ^{11}B (d, p) ^{12}B using a 1.5-MeV deuteron beam provided by the 5 MV Van de Graaff accelerator at Osaka University. The polarization was obtained when the produced nuclei were allowed to be ejected to an angle ranging from 32° to 48° relative to the incident beam. Recoiled ^{12}B nuclei were then implanted into a TiO_2 single crystal using their kinetic energy. An external magnetic field of H_0 = 5 kOe was applied anti-parallel to the direction of the polarization for maintaining the nuclear polarization and detecting the NMR. The polarization was monitored through the β-ray asymmetry detected by two sets of plastic-scintillation-counter telescopes placed along the external field H_0 such that one was above and the other below the TiO_2 crystal. The rf oscillating magnetic field H_1 was applied perpendicular to H_0 to induce magnetic transitions. The asymmetry change in the β-ray distribution was observed as a function of the rf at fixed H_0.

A typical β-NQR spectrum of ^{12}B in TiO_2 observed at room temperature is shown in Fig. 1 as a function of the separation frequency of the two transitions, which is defined as $\Delta\nu \approx 3ev_{ij}Q/2I(2I-1)h$ in first order perturbation. From the two resonance peaks in the spectrum in Fig. 1(a), (c-axis // H_0), we found two implantation sites with the relative populations (site 1) : (site 2) = 9 : 1. The sum of the polarizations detected at the two sites was that of the ^{12}B implanted in Pt metal, i. e., the polarization of ^{12}B as produced through the nuclear reaction was 100% kept in the TiO_2 crystal. In the spectrum shown in Fig. 1 (b), in which the crystal $\langle 110 \rangle$ axis was set parallel to H_0, we found a set of very close two peaks for ^{12}B in site 1 with a separation less than the frequency modulation width for β-NQR, which implies that the asymmetry parameter η is very small. Since it is not natural to assume a zero η because of the tetragonal structure of TiO_2, the present result is difficult to understand, and a detailed study is urged. From these β-NQR spectra, we extracted the electric field gradients (EFGs) of those two implantation sites as shown in Table 1. Here, the EFGs were determined using the well known Q moment of ^{12}B, $Q(^{12}\mathrm{B})$ = +(13.21 $\pm$ 0.26) mb [4, 5]. The signs of the EFGs were also determined from the asymmetric population of the single rf β-NMR spectra. The asymmetry of the NMR effect (asymmetry change) for the two peaks for each site in the NMR spectrum is caused by the positive initial spin alignment [6] of ^{12}B and the direction

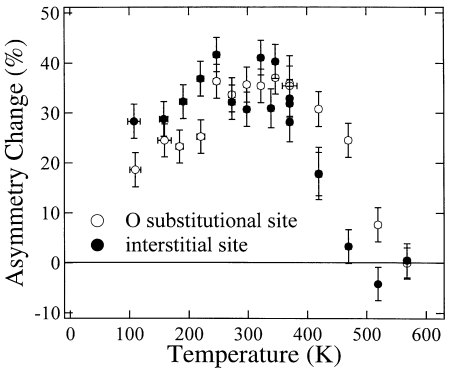

Fig. 3. Temperature dependence of the asymmetry change for ^{12}N in TiO_2. Open circles correspond to the O substitutional site and closed circles to the interstitial site.

of the polarization relative to H_0 besides the sign of the eqQ.

The amount of the preserved polarization was given from the β-ray asymmetry change as a function of temperature in the region from 110 K to 600 K as shown in Figure 2. The ^{12}B in Site 1 showed a clear temperature dependence. The polarization, although it was fully kept at 300 K, gradually decreased as the temperature decreased and reached 20% of the total at 110 K. At higher temperature there was a significant dip at around 400 K, and the polarization is recovered again at 600 K. On the other hand, the ^{12}B in Site 2 showed no peculiar temperature dependence. We confirmed that the EFGs did not change much in the present temperature range. We performed a similar measurement on ^{12}N ($I^\pi = 1^+$, $T_{1/2} = 11.0$ ms), which also had two implantation sites with the same population, i. e., the population ratio was 1:1 [1], as shown in Figure 3. A strong temperature dependence was observed for both sites. The decrease of the preserved polarization may be caused by either the decrease of the population in the sites or a change of the charge state which has to be explained by the electronic structure around the sites. A study to understand the present temperature dependences is now in progress.

3. Implantation Sites of ^{12}B in TiO_2

In order to identify the locations of ^{12}B in TiO_2, the EFGs were calculated from the charge density distribution of the system, which was determined self-consistently by use of the Korringa-Kohn-Rostoker (KKR) method with the local density approximation [7]. Details of the calculation were similar to the previous work [1]. We used a super cell which consists of two TiO_2 unit cells stacked along the c -axis. As the possible implantation sites, the Ti substitutional site, the O substitutional site and the octahedral like interstitial site were considered. The octahedral like interstitial site means a face center of the side of the tetragonal TiO_2 unit cell. To estimate the theoretical lattice relaxation around the impurity atom, the total energy of this super cell was calculated with a slight displacement of the neighboring atoms from their original locations. For the Ti substitutional site and the interstitial site, the neighboring six O atoms were relaxed and for the O substitutional site, the surrounding three Ti atoms were relaxed. The local lattice relaxation of about -8%, +5% and +39% were predicted for the Ti substitutional site, the O substitutional site and the interstitial site, respectively.

Table 2. Experimental and theoretical EFGs (in units of 10^{15} V/cm^2) at B impurities in TiO_2.

	V_{ii}// c-axis	V_{ii}//$\langle 110\rangle$	V_{ii}//$\langle 1\bar{1}0\rangle$
Experiment			
Site 1	+37.1 ± 0.5	–18.8 ± 0.4	–18.8 ± 0.4
Site 2	–150 ± 4	+185 ± 5	–35 ± 5
Theory			
Sub. Ti (B^{3+})	+63	–9	–54
Sub. O (B^-)	+10	+167	–177
Int. (B^-)	–179	+36	–143

The calculated EFGs are listed in Table 2 together with the experimental data. The assumed charge states are also shown. It seems that the experimental site 1 is the Ti substitutional site because the theory reproduced the sign of the main component of the EFGs. In the case of the O substitutional site, the sign of the component which is parallel to the c-axis also agrees with that of site 1, but the magnitude of the other components is too large. The small η detected by the experiment was not reproduced. The experimental site 2 is likely to be the octahedral interstitial site with the B^- charge state. The theory does not reproduce the magnitude of EFGs well for both cases. In order to understand these discrepancies further experimental and theoretical studies are continued.

4. Possible use of TiO_2 as a Spin Dewar

The results that TiO_2 keeps 100% polarization of ^{12}B and that 90% of ^{12}B occupy one site in TiO_2 imply that TiO_2 is a good catcher for spin control

Table 3. The preservation of nuclear polarization in TiO_2. Preservation ratio relative to the polarization as produced through nuclear reaction.

Nucleus ($T_{1/2}$)	Preservation ratio	Number of sites	T_1	Reference
^{8}Li (0.84 s)	30%			This work
^{8}B (0.77 s), ^{12}B (20 ms)	100%	2 (9:1)		This work
^{12}N (11 ms), ^{16}N (7.1 s)	100%	2 (1:1)	$>$ 10 s	[1]
^{13}O (8.6 ms), ^{19}O (27 s)	100%	2 (7:3)	$\sim$ 100 s	[2]
^{41}Sc (0.60 s)	100%	1		[3]

of B isotopes. Also, about 30% polarization of ^{8}Li ($I^{\pi} = 2^{+}$, $T_{1/2}$ = 838 ms) was maintained in TiO_2. Taking account of the results of ^{8}Li and ^{12}B as well as the previous data, it is concluded that TiO_2 is a good candidate for a Spin Dewar. The nuclei which keep their polarization in TiO_2 are summarized in Table 3. For the systematic study on the implantation of short-lived nuclei, we plan to investigate the implantation of various other β emitters such as ^{17,20}F, ^{21}Na and so on.

The final goal of the present study is to understand the mechanism of the preservation of the polarization in order to choose or design and control a perfect Spin Dewar for any specific nucleus. Studies for controlling the amount of preserved polarization and implantation sites freely are now in progress.

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