Quadrupole Couplings of N12 and B12 Implanted in Metal Single-Crystals

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Measurements have been made of the quadrupole couplings of $^{12}$N implanted in single crystals of Be and Mg and of $^{12}$B implanted in a single crystal of Zn. A comparison of the $^{12}$N couplings in Be and Mg suggests that (i) the final stopping sites of the implanted $^{12}$N ions are substitutional sites, i.e., the $^{12}$N ions occupy metal-ion lattice positions, and (ii) the $^{12}$N ions implanted in Be and Mg have the same charge state and quadrupole shielding factor. A procedure is outlined for deducing $Q(^{12}$N). The $^{12}$B couplings in Zn imply the existence of two inequivalent stopping sites. The problems encountered in deducing $Q(^{12}$B) from couplings in metal hexagonal crystals are discussed.

An experimental study of the quadrupole couplings of light $\beta$-unstable nuclei implanted in single crystals of hexagonal metals has recently been initiated. The purpose of the study is to investigate the possibility of extracting values for the nuclear quadrupole moments from the crystalline electric field gradients simply from values of the respective quadrupole couplings. This paper reports measurements of the quadrupole couplings of $^{12}$N implanted in single crystals of Be and Mg and of $^{12}$B implanted in a single crystal of Zn. A procedure is outlined for deducing $Q(^{12}$N), while the problems encountered in deducing $Q(^{12}$B) are discussed.

EXPERIMENTAL PROCEDURE

Polarized $^{12}$N and $^{12}$B recoils were produced at the Brookhaven National Laboratory 4-MeV Van de Graaff facility using the reactions $^{10}$B$(^3$He, n)$^{12}$N and $^{12}$B(d, p)$^{12}$B. The $\beta$-unstable nuclei decay according to the schemes $^{12}$N $\rightarrow ^{12}$C + $e^-$ + $\bar{\nu}$ and $^{12}$B $\rightarrow ^{12}$C + $e^-$ + $\bar{\nu}$. The half-lives of $^{12}$N and $^{12}$B are 11.0 and 20.4 msec, respectively, and their $\beta$ end-point energies are 16.4 and 13.4 MeV, respectively. The angular momenta and parities of the initial and final states are $1^+$ and $0^+$, respectively, in both decays. Hence both transitions are pure Gamov-Teller transitions; so any polarization of the $^{12}$N or $^{12}$B nuclei will result in an asymmetry in the emission directions of the decay betas. The polarized $^{12}$N and $^{12}$B nuclei were stopped in a single crystal of a hexagonal metal (Mg, Be, Zn), where the respective nuclear quadrupole moments interact with the crystalline electric field gradient. The advantages of single crystal over polycrystalline stopping materials have already been demonstrated in the case of the $^{12}$B quadrupole coupling in Be. The experimental technique employed in measuring the quadrupole couplings was simply resonant partial depolarization of the recoil nuclei and will be described briefly.

For the reaction $^{10}$B$(^3$He, n)$^{12}$N, a 100-$\mu$g/cm$^2$ layer of $^{10}$B was evaporated onto a thin Au foil and bombarded with a 3.4-MeV $^3$He beam. For the reaction $^{12}$B(d, p)$^{12}$B, a 60 $\mu$g/cm$^2$ layer of $^{12}$B was evaporated onto a thin Au foil and bombarded with a 1.6-MeV deuteron beam. The polarized recoiling $^{12}$N ($^{12}$B) nuclei were collimated so that the fraction centered about a laboratory recoil angle of 30° (44°) was allowed to strike one of the metal single crystals (Mg, Be, Zn). A large (~ kG) vertical magnetic holding field was applied normal to the reaction plane. The subsequent decay betas were detected by two coincidence telescopes, one positioned directly above and one directly below the metal stopping crystal. Each telescope contained one totally depleted silicon surface-barrier detector and one lithium-drifted silicon detector. The background counting rate was kept low by electrostatically chopping the $^3$He or deuteron beam and counting only when the beam was off. Typical beam times were 15 msec on, 20 msec off, for the $^{12}$N reaction and 20 msec on, 30 msec off, for the $^{12}$B reaction. Since the recoils were polarized perpendicular to the reaction plane, this polarization was detected by observing the asymmetry in the counting rates of the telescopes. Resonance lines were obtained by varying in discrete steps the frequency of a weak (~ G) oscillating horizontal magnetic field, while keeping the large vertical magnetic field constant. The partial destruction of the nuclear polarization at a resonance frequency was manifested as a change in the up-down $\beta$ asymmetry. The frequency changed approximately once every second; so random drifts in any part of the system were rapidly averaged out.

The lattice structure of the metal single crystals is hexagonal close packed and possesses electric field gradients at both lattice and interstitial positions. The interaction of the electric quadrupole moments of the implanted recoil nuclei with these electric field gradients can be treated, in the cases to be considered, as a perturbation of the Zeeman...
splitting. The energy of the nuclear magnetic sub-level $m$ is then:

$$E_m = -m \gamma h B + \frac{e^2 q Q}{4(2I - 1)} \frac{3 \cos^2 \theta - 1}{2} \frac{3m^2 - I(I + 1)}{2}$$

where $\gamma$ is the nuclear gyromagnetic ratio, $B$ is the magnitude of the large vertical magnetic field, $q$ is the electric field gradient at the nucleus, $Q$ is the nuclear electric quadrupole moment, $I$ is the nuclear spin ($I = 1$ for $^{12}$N and $^{12}$B), and $\theta$ is the angle between the direction of the electric field gradient, i.e., the crystalline $c$ axis, and the direction of the external magnetic field $B$. The frequencies of resonance lines then satisfy the relation

$$h \nu_{\text{resonance}} = \gamma h B \pm \frac{3 e^2 q Q}{8} (3 \cos^2 \theta - 1).$$

Instead of a single Zeeman resonance line, one obtains two quadrupole resonance lines equally split from the Zeeman frequency. The experimental procedure was to scan these quadrupole resonance lines for a series of different values of the angle $\theta$ corresponding to increasing quadrupole splittings.

$^{12}$N RESULTS

Figure 1 presents the results of resonance scans taken of $^{12}$N implanted in Be with $\theta = 70^\circ$ and $\theta = 90^\circ$, while Fig. 2 presents similar data for $^{12}$N implanted in Mg with $\theta = 90^\circ$ and $\theta = 0^\circ$. In both cases the frequency of the 2-G horizontal depolarizing field was varied in steps of 10 kHz, while the frequency at each step was modulated by $\pm 5$ kHz. Since the frequency of the sinusoidal modulating signal was 1.25 kHz, the modulation index was 4. The magnitude of the vertical magnetic holding field was obtained by measurement of the proton NMR frequency. For all $^{12}$N scans $|B|$ was 3574 $\pm$ 2 G.

The quadrupole splittings, $\frac{3}{2} \frac{e^2 q Q}{h} (3 \cos^2 \theta - 1)$, in Figs. 1 and 2 exhibit the proper dependence on the angle $\theta$ between the crystalline $c$ axis and the magnetic holding field $B$. The quadrupole splitting at $\theta = 90^\circ$ is 1.5 times the splitting at $\theta = 70^\circ$, while the splitting at $\theta = 0^\circ$ is 2 times the splitting at $\theta = 90^\circ$. The solid lines drawn through the data of Figs. 1 and 2 represent the results of a computer fit of the data to Gaussian resonance lines plus a constant background. Four constraints are imposed in this computer fit. First, the widths of the Gaussian resonance lines are constrained to be the same for scans in the same metal. Second, the ratio of the areas of the two resonance lines in each scan is constrained to be the same for all four resonance scans. Third, the positions of the Gaussian resonance lines are constrained to be equidistant from the Zeeman frequency (zero quadrupole splitting frequency). Fourth, this Zeeman frequency is then constrained to be the same for scans in the same metal. Thus the original 28
The Zeeman frequency of $^{12}$N in Be yielded by the computer fit is 1249 ± 3 kHz, while that of $^{12}$N in Mg is 1247 ± 2 kHz. Both values yield $\mu (^{12}\text{N}) = 0.458 \pm 0.001 \mu_N$, which is consistent with the more precise measurement made previously by the Osaka group.\(^\text{12}\)

Both the $^{25}\text{Mg}$ quadrupole coupling in magnesium metal and the $^{25}\text{Mg}$ nuclear quadrupole moment have been measured.\(^\text{5,6}\) Using the calculated value of the quadrupole shielding factor for Mg,\(^\text{7}\) one can deduce the value of the electric field gradient at a lattice position in magnesium metal. Similarly, from the measured value of the $^{9}\text{Be}$ nuclear quadrupole moment,\(^\text{8}\) the measured value of the $^{9}\text{Be}$ quadrupole coupling in beryllium metal,\(^\text{9}\) and the calculated value for the shielding factor for Be,\(^\text{10}\) one can deduce the electric field gradient at a lattice position in beryllium metal. Table II lists the results of these two calculations. It may now be noted that the ratio of the electric field gradient at a lattice position in magnesium metal to the gradient at a lattice position in beryllium metal should equal the ratio of the $^{12}$N coupling in Mg to the coupling in Be if the following two conditions hold: (i) the final stopping sites of the implanted $^{12}$N ions are substitutional sites in both Mg and Be, i.e., the $^{12}$N ions occupy metal ion lattice positions, and (ii) the $^{12}$N ions implanted in both Mg and Be have the same charge state and shielding factor. In fact, the agreement between the ratio of Table I and the ratio of Table II is striking. It should be noted that boron implanted in silicon at room temperature has been shown to occupy substitutional sites,\(^\text{11}\) rendering condition (i) at least plausible. Condition (ii) should hold unless the host metals Mg and Be interact very differently with the implanted $^{12}$N ion.

A procedure can be outlined for deducing the $^{12}$N nuclear quadrupole moment from the $^{12}$N quadrupole couplings in Be and Mg. First, conditions (i) and (ii) are assumed to hold. Under this assumption, the electric field gradients experienced by the $^{12}$N ions in Be and Mg are the quantities $q'$ calculated in Table II. Second, a charge state for the $^{12}$N ion must be selected, and the corresponding quadrupole shielding factor $\gamma$ must be calculated. Assuming a charge state of N$^+$ and choosing the $^{12}$N coupling in Be,

$$Q(^{12}\text{N}) = \frac{e^2 q'q/\hbar}{(1 - \gamma)(1 - \gamma^{{\text{Be}}})} = \frac{Q\text{calc}}{(1 - \gamma^{{\text{Be}}})} = \frac{Q\text{calc}}{1 - \gamma^{{\text{Be}}} \gamma}$$

where $\gamma = 0.22 \pm 0.06$ is an estimate by R. M. Sternheimer.\(^\text{13}\) The calculated value $Q(^{12}\text{N}) = 44 \pm 10$ mb is roughly an order of magnitude larger than a theoretical estimate made by Kurath,\(^\text{14}\)

$$Q(^{12}\text{N}) = 1.3 (e_p/e) + 12.0 (e_n/e) \text{ mb},$$

where $e_p$ and $e_n$ are the effective charges of the proton and neutron, respectively. Kurath's estimate is based on an $A = 12$ wave function [a linear combination of the lowest three $A = 12 (8 - 16)$ potential eigenfunctions] which yields theoretical values of the magnetic moments of $^{12}$B and $^{12}$N in reasonably good agreement with experimental values.

It should be stressed that a value for $Q(^{12}\text{N})$ calculated according to the above procedure can be no more reliable than the assumptions concerning the $^{12}$N stopping site and charge state. There-
fore additional experiments have been designed to provide an independent check on the validity of these assumptions.

13B RESULTS

Figure 3 presents the results of resonance scans taken of 12B implanted in Zn with 9 = 70°, 90°, 35°, and 0°. Since 12B is a negative β emitter while 12N is a positron emitter, resonance lines in Fig. 3 appear as dips in the beta asymmetry rather than as peaks as in the 12N scans. The magnitude of the horizontal rf field was approximately 4 G, while the frequency step interval and frequency modulation were identical to those employed in the 12N scans. Measurement of the proton NMR frequency implied a value for |I| of 1642.9 ± 0.5 G.

Using \( \mu(12B) = (1.00285 ± 0.00015) \mu_0 \) and |I| = 1642.9 ± 0.5 G, the Zeeman resonance frequency is calculated to be 1255.9 ± 0.4 kHz. Thus the two resonance lines in each scan appear to be split away from the Zeeman frequency in the same direction. In each scan the ratio of the quadrupole splittings, \( \frac{(e^2 qQ/\hbar)(3 \cos^2 \theta - 1)}{\cos \theta} \), of the two resonance lines is about 3 : 1.

One point needs a word of explanation. One would expect a single 12B stopping site to give rise to two resonance lines, equally split from the Zeeman frequency but in opposite directions. However, previous studies of 12B implanted in Be1 and Mg2 indicate that the populations of the 12B magnetic substates are such that a single stopping site yields two lines whose intensities are in the ratio 3 : 1. Hence in the present work only the stronger line is statistically evident.

The solid lines drawn through the data of Fig. 3 represent the results of a computer fit of the data to Gaussian resonance lines plus a constant background. The constraints imposed in this computer fit are identical to those imposed in the computer fit of the 12N data with one exception. The Zeeman frequency was fixed at 1255.9 ± 0.4 kHz for all four scans, ignoring any Knight shift for 12B in Zn. These constraints reduced the number of parameters from 28 to 16. The value of \( x^2 \) per degree of freedom and the values of the smaller quadrupole splittings and their standard deviations yielded by the computer fit are listed in Table III. The larger quadrupole coupling is calculated and listed by using the ratio of the larger to smaller couplings yielded by the computer fit.

In addition to the 12B quadrupole couplings in Zn reported in this paper, measurements have been made of the 12B couplings in single crystals of Be and Mg. Unfortunately, the magnitudes of the 12B quadrupole couplings in Be, Mg, and Zn do not exhibit the simple pattern of the 12N couplings in Be and Mg. For example, the ratio of the measured 12B coupling in Mg to the measured coupling in Be is 0.85 ± 0.01, not 0.22. The expected ratio of the quadrupole coupling of 12B in Be to the coupling in Zn and the expected ratio of the 12B coupling in Mg to that in Zn cannot be calculated by using the method outlined in Table II, since the quadrupole coupling of 67Zn in zinc metal has not been measured. Hence the questions of stopping site and charge state for 12B implanted in Be, Mg, and Zn require considerably more investigation.

CONCLUSIONS

A comparison of the quadrupole couplings of 12N implanted in single crystals of Be and Mg suggests...
that (i) the final stopping sites of the implanted $^{12}$N ions are substitutional sites in both Be and Mg, and (ii) the charge states and shielding factors of the implanted $^{12}$N ions are the same in both Be and Mg. If statements (i) and (ii) are assumed to be true and a charge state is selected for $^{12}$N implanted in Be and Mg, a procedure is available for deducing $Q_{eB} - N$.

Experiments have been designed to provide an independent check on the assumptions employed in this procedure.

Measurements of the quadrupole couplings of $^{12}$B implanted in a single crystal of Zn imply the existence of two inequivalent $^{12}$B stopping sites. More work is required before the $^{12}$B stopping sites can be identified and a reliable value of $Q(^{12}$B) can be deduced from $^{12}$B quadrupole couplings in hexagonal metal single crystals.

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