The Spin of Si^{29} and Mass Ratios of the Stable Si Isotopes*

R. L. WHITE AND C. H. TOWNES
Columbia University, New York, New York

(Received August 25, 1953)

The $J=0\rightarrow 1$ microwave transition of $Si^{29}D_3F$ has been examined under high resolution for hyperfine structure due to Si^{29}. No hyperfine structure was found, which sets an upper limit of 1×10^{-28} cm^2 to the quadrupole moment of Si^{29}. This is good evidence that the Si^{29} spin is 1/2. The measurement also allows an accurate determination of mass difference ratios for the Si isotopes.

CONFLICTING and somewhat inconclusive pieces of evidence on the spin of Si^{29} are already available from microwave and radiofrequency spectroscopy.[1–3] Although a recent angular correlation experiment[4] appears to provide evidence that the spin is 1/2, it seemed desirable to press the microwave measurements somewhat further than has previously been done.

If the spin of Si^{29} were greater than 1/2 and the nuclear quadrupole moment nonzero, the rotational microwave spectra of Si^{29} compounds would be expected to exhibit hyperfine structure arising from the coupling of the nuclear quadrupole moment to the molecular fields. The spectrum of $Si^{29}D_3F$ was examined with a high-resolution spectrometer in this laboratory, and no such hyperfine structure was found.

If the nuclear quadrupole coupling constant, eqQ, were small, an unresolved hyperfine structure appearing as line broadening might be expected. The $J=0\rightarrow 1$ transition of $Si^{29}D_3F$ has been measured to have a line width at half-intensity of 64 ± 6 kc/sec at $-78°$C in X-band waveguide at a pressure less than 10^{-3} mm. The same transition in $Si^{28}D_3F$ under identical conditions was observed to have a half-width of 63 ± 3 kc/sec. Si^{28} may be assumed to have spin 0, and hence no hyperfine structure. No evidence of line broadening in $Si^{29}D_3F$ is indicated. A quadrupole coupling constant eqQ of 50 kc/sec would give a line broadening of over 10 kc/sec. Hence if the spin of Si^{29} is not 1/2, its eqQ is less than 50 kc/sec in the compound examined. This is 2000 times smaller than the quadrupole coupling constant of Ge^{73} in the similar chemical compound GeH_3Cl.

One can calculate approximately what the maximum quadrupole moment of the Si^{29} nucleus might be consistent with the upper limit of 50 kc/sec on eqQ. This is accomplished by obtaining an approximate value of q, the gradient of the electric field along the molecular axis, from a knowledge of the bonding structure.[5] In the covalent structure of SiD_3F the three p orbitals are equally populated, hence are spherically symmetric and cannot contribute to q. In the ionic structure one p orbital is 75 percent vacated, assuming tetragonal hybridization. The Si—F bond might be expected to be 70 percent or more ionic on the basis of curves[6] of ionic character *versus* electronegativity differences, as well as from the bond length.[2] Let us assume then that the ionic structure is 80 percent important. Then the q in SiD_3F would be 60 percent of that in SiI. The q of SiI may be calculated by the usual method from the known doublet separation, 287 cm^{-1}, to be $q=8.5\times 10^{15}$ esu. Hence, for the partially ionic structure of SiD_3F, $q=5.1\times 10^{15}$ esu. This makes the quadrupole moment of Si^{29} a factor of 30 smaller than the smallest value observed for a nucleus (the deuteron) with a spin greater than 1/2, and a factor of several hundred smaller than the moments for neighboring nuclei. The quadrupole moment of Si^{29} would not be expected to be large from what is known of its structure. However, if the Si^{29} spin is not one half, its quadrupole moment should be roughly 100 times larger than the upper limit set by this experiment.

From the above, it may be seen that our failure to observe hyperfine structure in the $Si^{29}D_3F$ spectrum affords strong evidence that the spin of Si^{29} is 1/2.

The isotopic mass ratio for the stable silicon isotopes was also measured. The frequencies of the $J=0\rightarrow 1$ transition of the various isotopes were as follows:

$$Si^{30}D_3F = 24\,203.898\pm 0.002 \text{ Mc/sec},$$
$$Si^{29}D_3F = 24\,351.160\pm 0.003 \text{ Mc/sec},$$
$$Si^{28}D_3F = 24\,506.229\pm 0.003 \text{ Mc/sec}.$$

From these frequencies one computes the mass ratio:

$$\frac{M(30)-M(29)}{M(30)-M(28)} = 0.49934\pm 0.00003.$$

This compares extremely well with the measurement of Duckworth, Preston, and Woodcock,[7] from mass spectrographic data:

$$\frac{M(30)-M(29)}{M(30)-M(28)} = 0.49934\pm 0.00006.$$

* Work supported by the U. S. Atomic Energy Commission.
[1] Townes, Mays, and Dailey, Phys. Rev. **76**, 700 (1949).
[2] Sharbaugh, Thomas, and Pritchard, Phys. Rev. **78**, 64 (1950).
[3] R. H. Sands and G. E. Pake, Phys. Rev. **89**, 896 (1953).
[4] C. F. Black, Phys. Rev. **90**, 381 (1953).
[5] C. H. Townes and B. P. Dailey, J. Chem. Phys. **17**, 782 (1949).
[6] L. Pauling, *The Nature of the Chemical Bond* (Cornell University Press, Ithaca, 1945); C. H. Townes and B. P. Dailey, Phys. Rev. **78**, 346 (1950); W. Gordy, J. Chem. Phys. **19**, 792 (1951).
[7] Duckworth, Preston, and Woodcock, Phys. Rev. **79**, 188 (1950); H. E. Duckworth and R. S. Preston, Phys. Rev. **79**, 402 (1950).

It also compares well with an earlier microwave determination by Geschwind and Gunther-Mohr.[8]

$$\frac{M(30)-M(29)}{M(30)-M(28)}=0.49941\pm0.00005.$$

ACKNOWLEDGMENT

We are very grateful to Drs. Børge Bak and John Anderson of the University of Copenhagen for furnishing us the SiD_3F used in this investigation.

[8] S. Geschwind and G. R. Gunther-Mohr, Phys. Rev. **81**, 882 (1951).

PHYSICAL REVIEW VOLUME 92, NUMBER 5 DECEMBER 1, 1953

The Isomeric Transition of Pb^{207} as an Energy Standard in Beta Spectroscopy

DAVID E. ALBURGER*
Nobel Institute of Physics, Stockholm, Sweden
(Received August 24, 1953)

The K-conversion line of the 1.06-Mev isomeric transition following the electron-capture decay of Bi^{207} is suggested as a useful energy standard for beta-spectroscopy. While the 50-year half-life and 10 percent conversion coefficient are comparable to Cs^{137}, the 60 percent higher energy, larger percentage K to L momentum separation, and the possibility of making thin and more uniform sources may be advantageous. Precision measurements have been carried out in a double-focusing spectrometer by comparison with the 1.02-Mev electrons of RaC. The Pb^{207} K-line has a momentum of 4657.9 ± 1.0 gauss-cm from which an electron energy of 975.9 ± 0.3 kev and a transition energy of 1063.9 ± 0.3 kev are derived. Measurements have also been made on the K/L and $K/(L+M)$ ratios, which are 3.95 ± 0.25 and 3.00 ± 0.25 respectively. When compared with data on other $M4$ transitions, these results indicate that deviations from the empirical K/L curves take place at high Z. The usefulness of Bi^{207} as a test source for beta-gamma coincidence detectors is discussed.

INTRODUCTION

ABOVE 0.5 Mev the only generally useful internal-conversion reference standards which have been measured[1] up to the present with an accuracy of ~2 parts in 10^4 are the 3381.3 gauss-cm line of Cs^{137} and the 9986.7 gauss-cm line of ThX. Although the 1.33-Mev gamma ray of Co^{60} and two electron lines of RaC at 1.02 and 1.32 Mev have now been determined[2] within this accuracy, the Co conversion electrons are impractical to use because of low-conversion coefficient, small K to L separation, and poor specific activity, while the RaC has the difficulties of a 20-min half-life and a large beta-ray continuum under the lines.

Bi^{207} offers the possibility of partially filling in the wide gap between Cs^{137} and ThX, insofar as electron lines are concerned. This isotope, which decays by electron capture to Pb^{207}, was first reported[3] by Neumann and Perlman. They estimated the half-life as being approximately 50 years, and they found a number of internal-conversion lines which they measured in a shaped magnetic field beta-ray spectrometer at 1.5 percent resolution. By far the strongest line occurs at 975 kev, corresponding to a transition energy of 1063 kev. A 0.9-sec isomeric activity in lead, observed[4] somewhat earlier by Campbell and Goodrich, was interpreted[5] by Goldhaber and Sunyar as belonging to the 1.06-Mev gamma ray of Pb^{207}, which they then assigned as an $M4$ transition. Relative conversion coefficients of the 1.06- and 0.56-Mev gammas, derived by Grace and Prescott[6] and by Wapstra,[7] supported this assignment. Recently Friedlander *et al.* have shown[8] conclusively that the 0.9-sec activity belongs to Pb^{207}.

Although the decay scheme of Bi^{207} is not yet established, it is probable that the isomeric transition occurs in a fairly large percentage of the decays. The theoretical K-shell conversion coefficient of the isomer is 0.103, and one can expect a yield of conversion electrons quite comparable to that from a similar source strength of Cs^{137}. Because there is no electron continuum and the other lines in the Bi^{207} spectrum are relatively low in intensity and widely separated, the K-1064 line can be used with virtually no scattered background effects.

Aside from extending the energy range of electron reference lines by approximately 60 percent above Cs^{137}, the bismuth activity has several advantages over the latter. One of these is the larger percentage momentum separation between K and L lines resulting from the higher atomic number. In the case of cesium the K to L separation amounts to 3.5 percent in momentum. When

* National Science Foundation Fellow on leave from Brookhaven National Laboratory, now returned to latter address.
[1] Lindström, Siegbahn, and Wapstra, Proc. Phys. Soc. (London) **B66**, 54 (1953).
[2] Lindström, Hedgran, and Alburger, Phys. Rev. **89**, 1303 (1953).
[3] H. M. Neumann and I. Perlman, Phys. Rev. **81**, 958 (1951).
[4] C. Campbell and M. Goodrich, Phys. Rev. **78**, 640 (1950).
[5] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[6] M. A. Grace and J. R. Prescott, Phys. Rev. **84**, 1059 (1951).
[7] A. H. Wapstra, thesis, Amsterdam, 1953 (unpublished).
[8] Friedlander, Wilson, Ghiorso, and Perlman, Phys. Rev. **91**, 498 (1953).