



FIG. 1.

voltage gradients. The thickness constant is about twice that for the radial motion. After removing the polarization voltages of 30,000 volts per centimeter, the remanence polarization is enough to give a high coupling. Since the electrostriction constant is the slope of the electrostriction expansion-voltage curve, the total thickness and radial expansions can be obtained by integrating these curves, and the results are shown by the dotted lines of Fig. 1. The total increase in thickness is 5 parts in 10,000 up to a voltage gradient of 30,000 volts per centimeter.

The discovery of the thickness effect, and the ratio of its electrostrictive constant of about 2 to 1 to that for the radial mode, allows one to obtain a mechanism for this effect. Barium titanate above 120°C has a cubic structure, but at room temperature it becomes ferro-electric and has expanded by two-thirds of one percent³ along the direction of the ferro-electric axis, and contracted one-third of one percent along the other two axes. In a polycrystalline body, the domains are distributed in all directions. The application of a high direct-current field lines up a certain percentage of the domains along the direction of the field. Since if all the domains were lined up the thickness would increase by two-thirds percent or 6.6×10^{-3} , the data of Fig. 1 indicate that

$$\frac{5 \times 10^{-4}}{6.6 \times 10^{-3}} = .075, \quad (3)$$

or 7.5 percent of the domains are lined up by a field of 30,000 volts per centimeter. Since the small alternating voltage superimposed on the direct-current field is too small to cause domains as a whole to change orientation, its principal effect is probably the transferring of molecules from domains not lined up with the field to domains lined up with the field, across their common boundary, when the alternating current field adds to the direct-current field and the reverse process when the two are opposing. When more molecules are in the domains lined up with the field, the crystal increases in thickness by a certain percentage and decreases in a radial direction by about half as much. When fewer molecules are in the lined-up domains, the reverse process occurs. Since the transfer of molecules from one domain to another lags behind the applied field, this

is the cause of the dielectric hysteresis that lowers the Q of the vibrations to about 15. The transfer of molecules between adjacent domains can go on for an alternating field without a biasing polarization, but it cannot change the thickness unless a polarization bias exists.

¹ Shepard Roberts, "Dielectric and piezoelectric properties of barium titanate," *Phys. Rev.* **71**, 890 (1947).

² See, for example, W. P. Mason, "Properties of monoclinic crystals," *Phys. Rev.* **70**, 705 (1946).

³ H. D. Megaw, *Proc. Roy. Soc.* **189**, 261 (1947).

Angular Correlation of Successive Gamma-Ray Quanta

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THEORETICAL considerations^{1,2} predict a directional correlation of successive quanta of the form

$$W(\theta) = 1 + \sum_{i=1}^l A_i \cos^{2i}\theta$$

if $2l$ is the highest multipole order occurring. Attempts to demonstrate this effect experimentally have heretofore been inconclusive.³ We have studied coincidences between successive gamma-rays of Co^{60} , Sc^{46} , Y^{86} (106 day), and Cs^{134} at angles 180° and 90° between the counters, and found a pronounced anisotropy in the first two named and no correlation within the experimental error in the last two. Our results, together with the gamma-ray energies concerned, are shown in Table I. The quantity $(W(\pi))$

TABLE I. Anisotropy of gamma-ray coincidences.

Source	Co^{60}	Sc^{46}	Y^{86}	Cs^{134}
ϵ Gamma-rays	0.21 ± 0.025	0.20 ± 0.035	-0.05 ± 0.03	0.01 ± 0.04
Mev	1.1, 1.3	0.89, 1.12	0.91, 1.89	0.58, 0.78
Reference	4	5	6	7

$-W(\pi/2)/W(\pi/2)$ should be equal to $\sum A_i$. Our results for this quantity, denoted by ϵ in Table I, should be increased by about six percent, because of finite angular resolution, and by an unknown, but probably small, correction arising from scattering in the source. The angular resolution of about $\pm 15^\circ$ was determined experimentally by measurements on annihilation radiation. Possible important effects attributable to interatomic fields¹ were shown to be absent in the case of Co^{60} , which was studied in the form of solid chloride, in aqueous solution, and as gaseous cobalt-nitrosyl-carbonyl. The respective values of ϵ were 0.19 ± 0.04 , 0.20 ± 0.04 , and 0.22 ± 0.04 .

Spurious angular correlations can be caused by small contaminations by positron-emitting substances. This was ruled out in the case of our samples of cobalt and scandium by observing, with greatly improved angular resolution, coincidences from stronger samples of the same preparations. In this arrangement the effect of annihilation radiation would be greatly emphasized. It was found that no

observable part of our results was due to positron contamination.

An apparent anisotropy can be caused by secondary quanta scattered from one counter to the other. This effect was observed and appropriate lead shielding arranged to remove it. Its absence in the final experiments was shown by observations using Fe⁶⁰, which emits two non-coincident gamma-rays of 1.1 Mev and 1.3 Mev. The coincidence rate was equal to the expected chance and cosmic-ray background, and was independent of angle.

It is interesting to note that the observed values of ϵ are consistent, within the experimental errors, with the values of ΣA_i predicted,¹ if the gamma-rays of Co⁶⁰ and Sc⁴⁶ are quadrupole radiation and the angular momenta of the nuclear states are 4, 2 and 0 in order of decreasing excitation energy in both cases. These spin assignments are in good agreement with considerations based on selection rules.⁴ Further experiments are in progress. We wish to thank Mr. A. C. Miller for help in taking data. This work was supported in part by the Office of Naval Research.

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² G. Goertzel, Phys. Rev. **70**, 897 (1946).

³ Kikuchi, Watase, and Itoh, Zeits. f. Physik **119**, 185 (1942); R. Beringer, Phys. Rev. **63**, 23 (1943); W. M. Good, Phys. Rev. **70**, 978 (1946).

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⁶ J. R. Downing, M. Deutsch, and A. Roberts, Phys. Rev. **60**, 470 (1941).

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⁸ M. Deutsch, J. R. Downing, L. G. Elliott, J. W. Irvine, Jr., and A. Roberts, Phys. Rev. **62**, 3 (1942).

Preliminary Analysis of the Microwave Spectrum of SO₂*

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THE purpose of the present note is to indicate the progress made to date on the analysis of the rotational spectrum of SO₂ as observed in the frequency range 20,000–30,000 mc/sec.^{1,2}

Although thirty-two lines have been observed, approximate measurements of intensity and of temperature coefficients of intensity indicate that only five lines correspond to transitions of moderately low J of S³²O₂¹⁶ in the ground vibrational state. The remainder includes lines presumably caused by high J transitions, by transitions of molecules in higher vibrational states, and by isotopic species such as S³⁴O₂¹⁶, etc. In the analysis, SO₂ was considered to be a rigid asymmetric rotor, with effective moments of inertia replacing equilibrium moments of inertia.

From the Stark splitting of the lines at 23,413 and 29,460 mc/sec., it was possible to determine the values of the main quantum numbers, J , involved in the transitions. Without placing some limits on the range of values of the moments of inertia it seems quite difficult to pursue the

analysis beyond this point. If however, the S–O distance is limited to values between the single-bond and triple-bond distances determined from normal covalent radii (1.37Å–1.70Å), and the O–S–O angle is limited to 70–180°, a unique identification of the two resolvable lines can be made.

These transitions permitted a sufficiently accurate determination of the rotational constants to predict a line that could be easily identified with the line observed at 25,392 mc/sec. Using these three transitions (of lowest J), a readjustment of the rotational constants was made that permitted additional lines to be identified with little difficulty. After the identification was made, the rotational constants were evaluated by applying the method of least squares to the five lines listed in Table I.

Consistency of the identification was tested in two ways:

(1) Comparison of the observed quantity

$$\Delta^0 = I_c^0 - I_a^0 - I_b^0$$

with that calculated from the fundamental frequencies of S³²O₂¹⁶ with the aid of the formula given by Darling and Dennison.³ The results are

$$\begin{aligned}\Delta^0(\text{obs.}) &= 0.20 \times 10^{-40} \text{ g cm}^2, \\ \Delta^0(\text{calc.}) &= 0.24 \times 10^{-40} \text{ g cm}^2.\end{aligned}$$

(2) Comparison of the observed Stark effect with that computed from the rotational constants. The results for the component frequencies measured relative to the unsplit line are

$$\begin{aligned}[6_{1,6} \rightarrow 5_{2,4}] \text{ at } 23,412 \text{ mc/sec.} \\ \Delta\nu(\text{obs.}) &= (0.27 - 0.013M^2)e^2(\text{e.s.u.}), \\ \Delta\nu(\text{calc.}) &= (0.224 - 0.0110M^2)e^2(\text{e.s.u.}), \\ [3_{1,3} \rightarrow 4_{0,4}] \text{ at } 29,460 \text{ mc/sec.} \\ \Delta\nu(\text{obs.}) &= (0.31 - 0.034M^2)e^2(\text{e.s.u.}), \\ \Delta\nu(\text{calc.}) &= (0.294 - 0.0325M^2)e^2(\text{e.s.u.}).\end{aligned}$$

The effective moments of inertia based upon the above assignment are

$$\begin{aligned}I_c^0 &= 95.14 \times 10^{-40} \text{ g cm}^2, \\ I_b^0 &= 81.16 \times 10^{-40} \text{ g cm}^2, \\ I_a^0 &= 13.78 \times 10^{-40} \text{ g cm}^2.\end{aligned}$$

Since the effect of centrifugal distortion was neglected, the assignments given and the values of the effective moments of inertia must be taken to be tentative. Because of the influence of zero-point vibration and other factors, it is not possible to determine the molecular parameters directly from the effective moments of inertia. However,

TABLE I. Tentatively identified lines of the rotational spectrum of SO₂.

ν , mc/sec.	Tentative assignment	Rela- tive inten- sity	$I_{-70^\circ\text{C}}$ $I_{27^\circ\text{C}}$	Comments
20,420	13 _{2,12} → 12 _{3,9}	2	4	5 components observed
23,413	6 _{1,6} → 5 _{2,4}	1.5	5	
24,037	—	5	2	
24,083	9 _{1,9} → 8 _{2,6}	5	3	2 components observed
25,392	7 _{2,6} → 8 _{1,7}	10	3	
29,460	3 _{1,3} → 4 _{0,4}	—	—	