

proportional to $T^{-\frac{1}{2}}$. The interactions considered by Anderson,⁸ polarizability and rotational resonance, show a T^{-1} behavior. The statistical theory of Margenau⁹ is density, but not velocity, dependent and should exhibit a T^{-1} behavior also. Thus, the values found show that much of the broadening is due to the quadrupole

⁸ P. W. Anderson, Phys. Rev. **76**, 647 (1949); **80**, 511 (1950).

⁹ H. Margenau, Phys. Rev. **76**, 121 (1949).

interaction. The results of Beringer and Castle¹⁰ for strong-field Zeeman transitions, in agreement with the present work, show temperature dependences between $T^{-\frac{1}{2}}$ and T^{-1} . From the knowledge at hand it seems that the quadrupole-quadrupole and the rotational resonance interactions are both important mechanisms in the broadening of oxygen absorption spectra.

¹⁰ R. Beringer and J. G. Castle, Jr., Phys. Rev. **81**, 82 (1951).

Some Observations of Double- and Triple-Quantum Transitions*

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(Received November 16, 1953)

Data are presented which show that it is possible to observe transitions for which $\Delta J=0$, $\Delta m_J=\pm 2$ or ± 3 in a molecule characterized by a total angular J . It is also possible to observe transitions for which $\Delta F=0$, $\Delta m_F=\pm 2$ or ± 3 in an atom characterized by a total angular momentum F . In each case the process occurs by the absorption or stimulated emission of two or three equienergetic quanta.

CERTAIN experiments, to be described elsewhere, require the measurement of the frequencies of the lines resulting from the transitions $\Delta J=0$, $\Delta m_J=\pm 1$ in the ground, $^3\Sigma$, state of the O_2 molecule for several values of K , the rotational angular momentum. At low values of the magnetic field, all lines for a particular J value coincide, but at magnetic fields sufficiently high to require terms, quadratic in the field, for the representation of the energy levels, $2J$ lines may be expected to appear for each J value.

The observation of the indicated spectrum presents considerable difficulty. A molecular beam of O_2 , issuing from a source of $77^\circ K$ was detected on a Pirani gauge, and the total deflection of an indicating galvanometer was small. In addition, even at $77^\circ K$, a significant number of rotational levels is occupied and the population of each state is small. For example, the

total population for $K=1$ is about 15 percent and the population of each J , m_J state is about 1.7 percent. Thus 3.3 percent of the particles in the beam may be involved in a particular transition. However, as Torrey¹ points out, the velocity distribution of the molecules has the effect of permitting a maximum of 0.766 of the particles to undergo a transition and the maximum effect which would appear is then about 2.6 percent of the refocused beam. It is to be noted that the effect to be observed as a consequence of a transition will generally be less than indicated as a consequence of an imperfect adjustment of the rf amplitude which induces the transitions and a limited deflecting power of the apparatus in which the transitions are observed.

In Fig. 1 is shown the spectrum resulting from the transitions which are nominally characterized by $\Delta m_J=\pm 1$ in the state $K=1$, $J=2$. It is at once seen that seven lines appear instead of the four predicted. Five of the lines (*A*, *B*, *D*, *F*, *G*) have an intensity of the order of that predicted for a single line, while two of the lines (*C*, *E*) have an intensity far greater than that predicted, beyond the range of experimental uncertainty.

We ascribe the observed spectrum to three processes: (1) the absorption or emission of a single quantum to give a transition for which $\Delta m_J=\pm 1$ (the lines *A* and *G* are produced by this process exclusively); (2) the absorption or emission of two equienergetic quanta to give a transition for which $\Delta m_J=\pm 2$ (the lines *B*, *D*, and *F* arise from this process); (3) the absorption or emission of three equienergetic quanta to give a transition for which $\Delta m_J=\pm 3$. The lines *C* and *E* result from

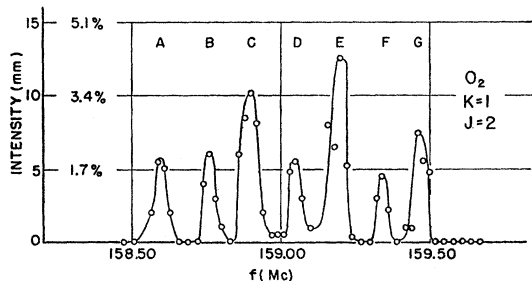


FIG. 1. The spectrum resulting from the transitions $\Delta J=0$, $\Delta m_J=\pm 1$ in the $^3\Sigma$ state of O_2 and for $K=1$, $J=2$ at a field of about 113 gauss. The intensity of the lines is given both as a fraction of the total beam and in absolute magnitude as measured on the apparatus.

* This research has been supported in part by the U. S. Office of Naval Research.

¹ H. C. Torrey, Phys. Rev. **59**, 293 (1941).

the superposition of the single-quantum transitions (1) and the three-quantum transitions (3). To the extent to which the energies of the levels can be represented by terms which are linear and quadratic in the field,

$$W = am_J H + bm_J^2 H^2;$$

the lines are all equally spaced and the frequencies of the triple transitions are exactly coincident with those of the single transitions. It should be noted that the triple transition at a particular frequency involves neither of the levels in which the single transition at the same frequency arises, and hence a total intensity, approximately twice that of the single transition, may be expected when the triple transition occurs. Finally it should be noted that a four-quantum transition may contribute to the intensity of the line *D*.

That the structure of the spectrum does not result from an inhomogeneity of the magnetic field is indicated by observations on the lines arising in $K=1, J=1$. The two lines had, at the same magnetic field as that used for the observations in Fig. 1, a mean frequency of about 164 Mc/sec but the separation between them was about 1.50 Mc/sec. The lines had no close satellites, but by an increase in the rf amplitude a new line arising

TABLE I. Notation used in describing transition in the spectrum $\Delta F=0, \Delta m_F=\pm 1$, of K^{39} .

Line designation	Transition ($F, m \leftrightarrow F, m'$)
α	(2, $2 \leftrightarrow 2, 1$)
β	(2, $1 \leftrightarrow 1, 0$), (1, $1 \leftrightarrow 1, 0$)
γ	(2, $0 \leftrightarrow 2, -1$), (1, $0 \leftrightarrow 1, -1$)
δ	(2, $-1 \leftrightarrow 2, -2$)

from the double-quantum transition appeared at the mean frequency of the two single-quantum transitions. The intensity of the new line was markedly sensitive to rf amplitude, and the line was unobservable when the single-quantum transitions still had a large intensity. The relative intensities of the lines arising in $J=2$ could also be modified by a change in the rf amplitude, where the lines *B*, *D*, and *F* were preferentially suppressed with decreasing amplitude.

An attempt was made to observe the transitions *B*, *D*, *F* as a single-quantum transition for which $\Delta m_J = \pm 2$ by application of an rf field of twice the frequency shown in Fig. 1. No evidence for the occurrence of the transition could be found and the possibility that a second harmonic of the oscillator produced the observed transition is thus excluded.

Since the observation and detailed analyses of the lines is made excessively difficult in the case of O_2 by the small beam intensity, further observations were made on the spectrum associated with the transitions $\Delta F=0, \Delta m_F = \pm 1$ in K^{39} . The spectrum was observed with an apparatus previously described.² In this ap-

² P. Kusch and H. M. Foley, Phys. Rev. **74**, 250 (1948); H. Tansb and P. Kusch, Phys. Rev. **75**, 1481 (1949).

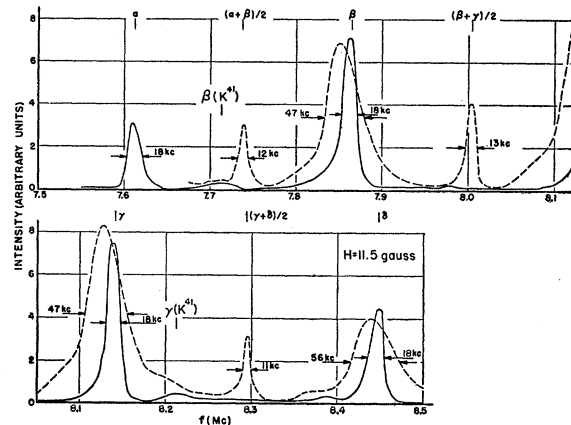


FIG. 2. The spectrum resulting from the transitions $\Delta F=0, \Delta m_F = \pm 1$ for K^{39} . The solid line shows the spectrum for low rf amplitude and the dotted line for high rf amplitude.

paratus the deflecting fields are very long; in addition the ratio of the gradient of the magnetic field and the field is high $[(\partial H/\partial Z)/H \cong 8]$. Accordingly, the field necessary to give particles with a moment of the order of a Bohr magneton an adequate deflection is sufficiently low so that potassium atoms are characterized by the quantum numbers F and m_F . In this quantization, for an atom with a spin of $\frac{3}{2}$ and in the $^2S_{3/2}$ state, atoms which have the same F but differ by 1 in m_F have moments which differ by $\frac{1}{2}\mu_0$. Hence it is possible to observe all transitions characterized by $\Delta F=0, \Delta m_F = \pm 1$. The length of the rf transition field was 2.5 cm. Since $I = \frac{3}{2}$ and the atom occurs in the $^2S_{3/2}$ state, $F=2$ or 1, and four lines appear in the state $F=2$ and two in $F=1$. Each of the two lines arising in $F=1$ forms a close doublet of separation $2g_I\mu_0 H/h$ with one of the lines in $F=2$. In all cases of interest here the doublet separation is much smaller than the natural width of the lines. The notation shown in Table I is used. The double-quantum transitions are designated as $(\alpha+\beta)/2$, etc.

In Fig. 2 is shown the observed spectrum at a field of about 11.5 gauss for a low rf amplitude (solid line) at which the single-quantum transitions are characterized by their natural half-widths (~ 18 kc/sec) and the spectrum for a high rf amplitude (dotted line) at which the double-quantum transitions have become prominent. In Table II are given the frequencies of the single-quantum transitions at both low and high rf amplitudes and of the double-quantum transitions at high rf amplitude. It is possible to choose a value of 0.069456 for the field parameter x ,

$$x = (g_J - g_I)\mu_0 H/h\Delta\nu,$$

such that the calculated frequencies of the single-quantum transitions at low amplitude and of the double-quantum transitions at high amplitude agree, within experimental error, with the observed frequencies. It thus appears that the double-quantum transitions

TABLE II. The observed and calculated frequencies of lines in the spectrum of K^{39} for both low and high rf amplitudes.

line	Low ampl.	
	obs $f(\text{Mc/sec})$	calc $f(\text{Mc/sec})$
α	7.612	7.612
β	7.862	7.864
γ	8.139	8.141
δ	8.449	8.447
	High ampl.	
$(\alpha+\beta)/2$	7.739	7.737
β	7.852	7.864
$(\beta+\gamma)/2$	8.004	8.003
γ	8.129	8.141
$(\gamma+\delta)/2$	8.295	8.293
δ	8.440	8.447

do, in fact, occur at a frequency which is determined from the elementary theory of the energy levels at a known magnetic field. It is evident, however, both from Fig. 2 and the data of Table II that the single-quantum transitions at high rf amplitude are shifted downwards in frequency. The data in the table are given in the sequence in which observations were made. It is very improbable that the indicated discrepancies can be ascribed to a drift in the magnetic field. It is not clear from experimental data alone that the shift is wholly related to the increased rf amplitude, since data exist in which lines are badly distorted at high rf amplitude because of inhomogeneities in the magnetic field in which transitions occur.

The half-widths of all the single-quantum lines agree closely with the predicted width of 18 kc/sec. The double-quantum lines are conspicuously narrower, of the order of 12 kc/sec. Substantially similar widths have been observed at a field of about 20 gauss, where, however, the existence of a shift cannot be established because of a field drift.

Observations have also been made of the three-quantum transition which can, of course, occur only in the upper ($F=2$) state. It can be shown from the usual expressions for the energy levels of the states that the frequency f_t of the triple transition and the fre-

quency f_s of the single transition are related by

$$\delta = |f_t - f_s| = x^3 \Delta\nu / 16$$

to terms in H^3 . Since $\Delta\nu = 461.7$ Mc/sec, $\delta \cong 100$ kc/sec at $x \cong 0.15$, which corresponds to $H = 25$ gauss. The lines should be resolved under these conditions. Figure 3 shows the three transitions $(\alpha+\beta)/2, \beta$, and $(\alpha+\beta+\gamma)/3$. The rf amplitude was so adjusted that the triple transition was of less than the maximum intensity. A small increase in rf amplitude increased the intensity of the line to about twice the indicated intensity with no significant increase in half-width. The dashes indicate a set of values for the line frequencies calculated directly from the energy level expressions where the frequency of the triple transition is used to determine the field. It is seen that the frequencies of the lines $(\alpha+\beta+\gamma)/3$ and $(\alpha+\beta)/2$ are very nearly consistent, while the line β is shifted to lower frequencies by about 50 kc/sec.

The observations recorded in Fig. 3 have been repeated. In Table III are recorded the observed frequencies, in the order of measurement, of the lines β , $(\alpha+\beta)/2$, and $(\alpha+\beta+\gamma)/3$. The first and last

TABLE III. The frequencies of several lines in the spectrum of K^{39} .

Line	Rf ampl.	Obs $f(\text{Mc/sec})$	Calc $f(\text{Mc/sec})$	Half-width kc/sec
β	Low	17.168	17.168 (Assumed)	19.5
$(\alpha+\beta+\gamma)/3$	High	17.252	17.251	20
β	High	17.118	17.168	66
$(\alpha+\beta)/2$	High	16.610	16.612	40
β	Low	17.167	17.168 (Assumed)	27

measurement indicate the absence of any significant drift in the magnetic field. The data indicate that the double- and triple-quantum transitions at high amplitude have frequencies consistent with the single-quantum transition at low amplitude but that the single-quantum transition at high amplitude is shifted to lower frequencies by 50 kc/sec.

It has not been possible to postulate any instrumental effect or observational procedure which would give rise to the indicated shifts. Particularly impressive are the widths of the single-quantum lines at low rf amplitude, which seem to preclude the existence of large inhomogeneities in the region of the magnet in which the transitions occurred. It is also remarkable, in Fig. 3, that the double-quantum transition with a width of 70 kc/sec is very nearly of the correct frequency relative to that of the triple-quantum transition, while the single-quantum transition with a width of 95 kc/sec is shifted to lower frequencies by 50 kc/sec. Nevertheless, the reality of shifts as atomic phenomena, not related to malfunctioning of the apparatus is not clearly established. The data here presented on the spectrum of potassium were made with a transition field not designed to produce a low and highly homogeneous

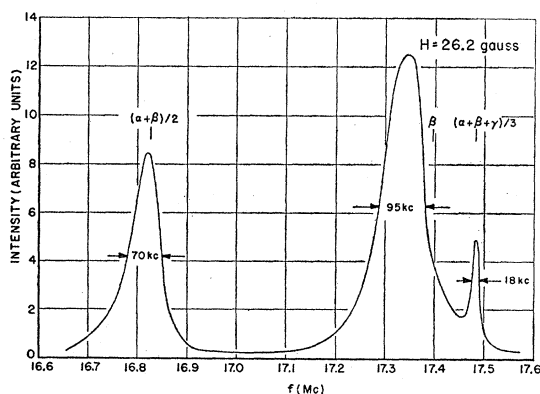


FIG. 3. A portion of the spectrum of K^{39} observed at a high rf amplitude.

magnetic field and which, moreover, has a history which suggests the existence of a considerable and variable inhomogeneity. By the criteria of the data it appears that, by good fortune, the data were taken in a section of the magnet which was homogeneous at the time of the observations. Extensive efforts to obtain lines as narrow as those whose observation is here reported by subjecting the magnet to various magnetizing and demagnetizing procedures have been unsuccessful; ultimately it appeared that the efforts were misdirected, in the sense that even a successful effort would be a rare accident and would not have any inherent criterion of the quality of the field external to the data obtained by use of the field. Apparatus is now being constructed in which the field will have much better inherent properties.³

Hughes and Grabner report the occurrence of certain double-quantum transitions in the radio-frequency spectrum of RbF observed by the electric resonance method. These occur between two levels between which a single-quantum transition is also allowed in the presence of a static electric field. They correspond to the absorption or emission of two quanta, which may be equienergetic, whose total energy is equal to that

³ V. Hughes and L. Grabner, *Phys. Rev.* **79**, 314 (1950); **79**, 829 (1950); **82**, 561 (1951).

of the single quantum ordinarily absorbed or emitted. The double-quantum transitions occur when the amplitude of the field and the static electric field are of comparable magnitude. The phenomenon is different from that here reported where the double- and triple-quantum transitions occur between states between which a single-quantum transition cannot ordinarily occur. The present effects depend on states intermediate to the terminal states of the multiple-quantum transition; the position of the intermediate states relative to equally spaced intervals between the terminal states strongly affects the probability of the multiple-quantum transition for any rf amplitude. On both experimental and theoretical grounds, the transitions here reported can occur even when the radio-frequency magnetic field has an amplitude small compared to the static field.

A discussion of the theory of multiple-quantum transitions and its application to the present data will be given by Mr. R. Salwen in a forthcoming paper. The theory predicts line widths which are in satisfactory agreement with those here reported. However, no satisfactory explanation of shifts of the order of those presently observed has been obtained.

I am indebted to Mr. Joseph Hendrie and to Mr. A. G. Prodel for their aid in taking the data of these experiments.

A Determination of the Energy of Antimony-Beryllium Photoneutrons

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(Received November 27, 1953)

The effective energy of Sb-Be photoneutrons has been measured by comparison of the transmission cross sections of five elements for the photoneutrons with previously measured cross-section values over a range of energies. The result placed the energy close to 25 kev.

INTRODUCTION

INTENSE gamma radiation from Sb¹²⁴ falling upon Be⁹ provides a copious source of nearly monoenergetic neutrons,¹ and has been used extensively for activation cross-section determinations.^{2,3} Degradation by various scattering processes in any source large enough to be useful poses the main difficulty in assigning the proper energy to the Sb-Be photoneutrons.⁴ Several measurements by various methods and with different source sizes have been made,^{1,5,6} with values

being quoted from 25 to 35 kilovolts. The present work concerns the possibility of narrowing the limits of error in the stated energy by a comparison of transmission cross sections for Sb-Be photoneutrons with transmission cross sections measured with the Argonne electrostatic generator in the 20 to 40 kilovolt region.

The specific purpose of this research was to assign a more definite value to the energy of the neutrons from that photoneutron source used by Hummel, Hamermesh, and Kimball^{2,3} in their activation cross-section work. A similar source was also used by others^{1,4,7} in various experiments involving cross sections, energy, and abundance of Sb-Be photoneutrons. The energy value determined in this experiment will thus provide cross checks on these earlier data.

¹ A. Wattenberg, *Phys. Rev.* **71**, 497 (1947).

² V. Hummel and B. Hamermesh, *Phys. Rev.* **82**, 67 (1951).

³ C. Kimball and B. Hamermesh, *Phys. Rev.* **89**, 1306 (1953).

⁴ Fields, Russell, Sachs, and Wattenberg, *Phys. Rev.* **71**, 508 (1947).

⁵ G. S. Klaiber and G. Scharff-Goldhaber, *Phys. Rev.* **67**, 733 (1942).

⁶ D. Hughes and C. Egler, *Phys. Rev.* **72**, 902 (1947).

⁷ Russell, Sachs, Wattenberg, and Fields, *Phys. Rev.* **73**, 545 (1948).