THE EFFECT OF HYPERFINE STRUCTURE DUE TO NUCLEAR SPIN ON POLARIZATION OF RESONANCE RADIATION

By A. Ellett
Department of Physics, University of Iowa

(Received February 6, 1930)

Abstract

The existence of hyperfine structure must be taken into account in calculations of the polarization of resonance radiation based upon Heinsenberg's extension of the principle of spectroscopic stability. Where the hyperfine structures are due to the existence of a nuclear moment their effect upon the polarization of resonance radiation may be calculated. If the nuclear moment of the thallium atom is $\frac{1}{2}$ (in units of $h/2\pi$) as Schüler and Brück suppose the λλ3776 and 5350 lines should show no polarization, while λ2768 should show 33.3 to 35.1 percent parallel and λ3530 41.8 to 48.8 percent perpendicular to the electric vector of a plane polarized exciting beam. Sodium resonance radiation excited by plane polarized D_1 and D_2 lines should show 33.3 percent polarization if the nuclear moment is $\frac{1}{2}$ and 16.6 percent if it is 1. The latter value agrees well with 16.3 observed by the writer, but observations on band spectra seem to indicate a higher nuclear moment, according to F. W. Loomis and R. S. Mulliken (Verbal communication to the writer.)

THE success of Back and Goudsmidt[1] in accounting for the hyperfine structure and Zeeman effect of the Bismuth arc spectrum by ascribing a mechanical and magnetic moment to the nucleus of the atom has led to attempts to explain hyperfine structure found in various other spectra in the same manner. Schüler and Brück[2] have shown that the hyperfine structures of the TlI spectrum are probably due to a nuclear moment of $\frac{1}{2}$ (in units of $h/2\pi$) and McLennan, McLay, and Crawford[3] have shown that their observations on Tl II lend support to this hypothesis. More recently Schüler and Brück[4] have had some success in unraveling the hyperfine structures of Cd I by supposing that this spectrum is really two superimposed—one without hyperfine structure due to the cadmium isotopes of even atomic number which are supposed to have no nuclear moment, and another with structure due to a nuclear moment ascribed to the odd isotopes.[5] The idea of nuclear spin has been used also by Jackson[6] and by Hargreaves[7] to account for Jackson's observations on the hyperfine structure of Cs I.

[1] Back and Goudsmidt, Zeits. f. Physik **43**, 321 (1927); **47**, 174 (1928). Pauli, Naturwiss. **12**, 741 (1924) suggested the idea of nuclear spin, but it was not successfully applied to the explanation of hyperfine structures until the paper of Back and Goudsmidt.

[2] Schüler and Brück, Zeits. f. Physik **55**, 575 (1929).

[3] McLennan, McLay and Crawford, Proc. R S **A125**, 570 (1929).

[4] Schüler and Brück, Zeits. f. Physik **56**, 291 (1929).

[5] Goudsmidt, Naturwiss. **41**, 805 (1929).

[6] Jackson, Proc. Roy. Soc. **A121**, 805 (1929).

[7] Hargreaves, Proc. Roy Soc. **A124**, 568 (1929).

That the existence of hyperfine structures must be taken into account in any comparison of theory and experiment on the polarization of resonance radiation or of radiation excited by electron impact is obvious for the calculation of the polarization in the absence of a magnetic field, based on the idea of spectroscopic stability,[8] involves the relative intensities of the Zeeman components of the line in *very weak* fields.

All previous calculations of the polarization of resonance radiation have been carried out without taking into account the existence of hyperfine structure. The Zeeman patterns and transition probabilities used have been those calculated for an atomic model without nuclear spin. These intensities are, at least to a first approximation, those which will obtain when the Paschen Back effect of the *hyperfine structure* is complete. There is no reason to expect that calculations made on this basis will show good agreement with experiment and in fact the agreement in general is poor. That hyperfine-structure is responsible for the 80 percent polarization of the λ2537 A. U. resonance radiation of mercury has been shown experimentally by MacNair and the writer.[9]

It is not likely that the hyperfine structure of this line is to be accounted for on the hypothesis of nuclear spin alone but in the case of thallium and perhaps sodium the situation is different, and it is interesting to see whether taking into account the nuclear spin will lead to better agreement between theory and experiment.

The present calculations are based on the assumption that the coupling between i, J, and f is of the same character as that between L, S and J in alkali spectra. That is, it is supposed that i, the nuclear moment, combines vectorially with J, the net moment due to the remainder of the atom to give a resultant f in the same way that L and S combine to give a resultant J. This is the assumption used by Back and Goudsmidt[1] and by Schüler and Brück.[2,4] The relative intensities of the hyperfine lines making up a hyperfine structure pattern (which itself appears as a 'line' save under high revolving power) may be calculated by formulas given by Hill[10] and by Hargreaves.[7] The relative intensities of the Zeeman components of the hyperfine lines may be calculated by the usual Kronig-Hönl formulas.[11] The polarization to be expected in any specified experiment is then calculated by making use of a consequence of the principle of spectroscopic stability first enunciated by Heisenberg.[8] This extension of the principle of spectroscopic stability may be applied in any case where the experimental situation presents an axis of symmetry, and it states specifically that the introduction of a magnetic field parallel to this axis will not alter the relative probabilities of transitions giving rise to radiation polarized respectively parallel or perpendicular to this symmetry axis. The field is introduced merely to remove the degeneracy which exists in the absence of any field. That is, its purpose is to render

[8] Bohr, Naturwiss. **12**, 1115 (1924); Heisenberg, Zeits. f. Physik **31**, 617 (1926).
[9] MacNair and Ellett, Phys. Rev. **31**, 180 (1928).
[10] Hill Proc. Nat. Acad. Sci. **15**, 779 (1929).
[11] Kronig, Zeits. f. Physik **31**, 885 (1925); Hönl, Zeits. f. Physik **31**, 340 (1925).

distinct the various magnetic energy levels which coalesce in the absence of a field so that the relative probabilities of transitions between these levels may be calculated. While this hypothetical field must be a strong field in the sense that the degeneracy is removed completely it must at the same time be a weak field so far as the production of Paschen Back effect of the hyperfine structure is concerned. The justification of such procedure then requires that the separation between the various hyperfine structure components of a line should be large compared to the intrinsic breadth of these components. This intrinsic breadth has nothing to do with the breadth of lines as observed in high resolving power spectroscopes; the latter is due to Doppler effect and pressure broadening (collisions) etc. We refer here to the inherent breadth of the line occasioned by imperfect definition of energy levels resulting from the finite life of stationary states.

To calculate the polarization of resonance radiation excited by plane polarized exciting light in the absence of a magnetic field we must imagine a field introduced, its direction being that of the symmetry axis (here the electric vector of the exciting light) so that we may calculate the relative numbers of transitions to the several Zeeman levels into which the upper states involved are split. Then having calculated the relative populations of these levels we determine the relative numbers of return transitions giving rise to radiation polarized parallel or perpendicular to the field. From this we get the polarization of the reemitted radiation and are assured that the value so calculated is that which will obtain when the field is removed.

THALLIUM

The thallium lines arise from transitions shown in the Grotrian diagram, Fig. 1a. Each of the levels shown is split into two when a nuclear moment of $\frac{1}{2}$ is taken into account, and these with the appropriate values of the fine quantum number f, and the permitted transitions are given in Figs. 1b and 2. The values of the transition probabilities are given by Hargreaves[7]—or

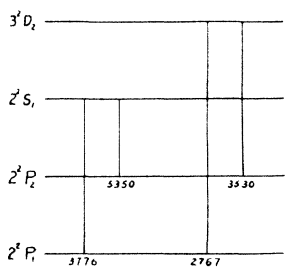

Fig. 1a. Grotrian diagram for thallium.

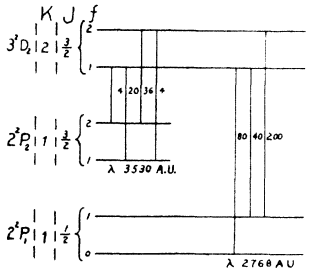

Fig. 1b. Permitted transitions in thallium.

they may be readily computed from the formulas given by Hill.[9] From the figures it is evident that excitation by λ3776 A. U. will give rise to the emission of this line and also λ5350 A. U. while absorbtion of λ2768 A. U. excites λ3530 A. U. in addition to itself. In Fig. 2 are shown the Zeeman levels and

transitions involved in the emission of λλ3776 and 5350 A. U. Excitation by plane polarized light will excite atoms to those upper energy levels reached by those transitions of λ3776 A. U. marked || (parallel) in the figure, the relative numbers of excited atoms in the various upper states depending upon the values of the parallel transitions reaching it and upon the intensity of the incident light of wave-length appropriate to produce these transitions. The question of relative intensities in the exciting radiation need not bother us here however, for no matter what the relative populations of the upper levels the reemitted radiation of each of the aggregates of hyperfine lines composing λλ3776 and 5350 A. U. will be unpolarized. That is to say, in each of these aggregates from *each* upper level the chance of returning by a perpen-

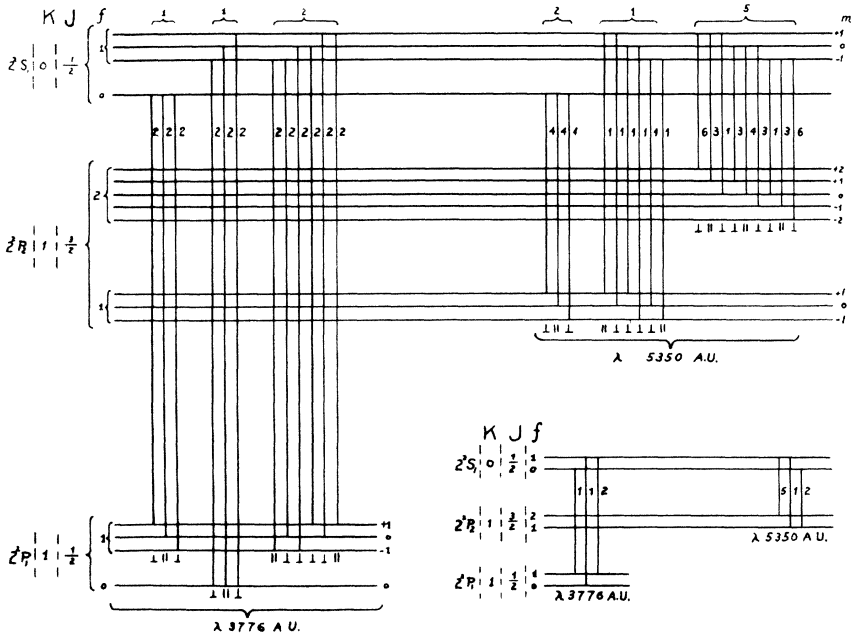

Fig. 2. Permitted transitions in thallium.

dicular transition is twice that of returning by a parallel transition. This leads to no net polarization either parallel or perpendicular to the electric vector of the exciting light—for parallel transitions produce such a distribution of the reemitted radiation as would be produced by a linear oscillator parallel to the electric vector of the exciting light (and also parallel to our imaginary magnetic field) while perpendicular transitions give rise to a distribution such as would be occasioned by a circular oscillator in a plane perperdicular to this.

Figure 3 shows the levels involved in absorption and emission of λλ2768 and 3530 A. U. Here the relative population of the upper levels is of some consequence and it becomes necessary to make some assumption regarding the distribution of intensity over the hyperfine structure components of

λ2768 A. U. Two cases will be considered —A— that of uniform intensity over the entire absorption pattern, a situation perhaps realizable with a very hot source emitting greatly broadened lines, and —B— that of the theoretical intensity ratios,—such as might be realized with a source run at a temperature low enough so that Doppler broadening will cause no overlap of

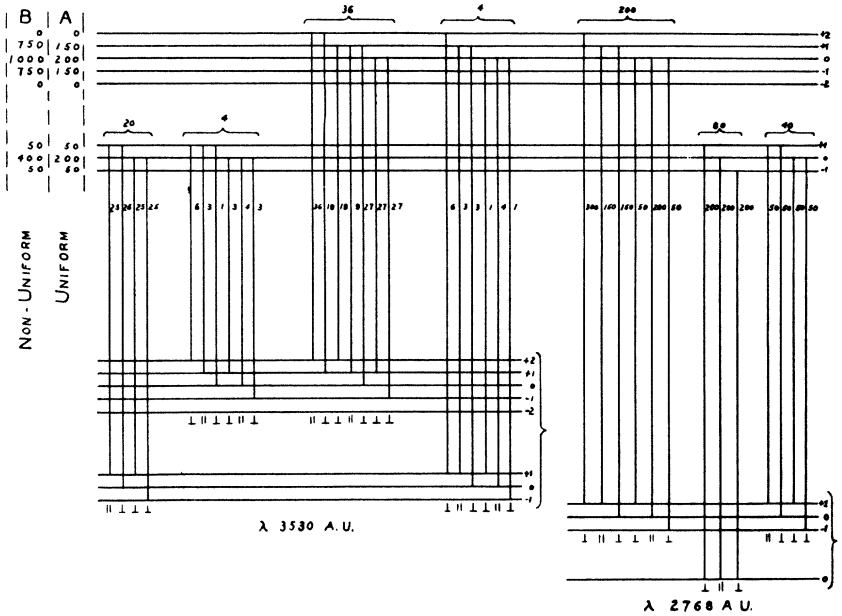

Fig. 3. Levels involved in absorption and emission of λλ2768 and 3530.

hyperfine structure components, and with the density of thallium vapor low enough to eliminate self reversal. In column A of Fig. 3 are given the relative populations of the upper levels for case A and in column B for case B. Table I summarizes the results for these four lines.

TABLE I.

Nuclear moment i	0	1/2		observed
		A	*B*	
2768	60 ∥	33.3 ∥	35.1 ∥	35 ∥ (500°C) 55 ∥ (470°C)[12a]
3530	75 ⊥	41.8 ⊥	48.8 ⊥	30 ⊥ (500°C) 60 ⊥ (470°C)[12a]
3776	0	0	0	0[12b]
5350	0	0	0	0[12b]

Observed values[12a,b] are given in column five, while the values to be expected for an atom without nuclear moment are given in column two.

More recently Schüler and Brück[13] have advanced the hypothesis that in

[12a] Gülke, Zeits. f. Physik **56**, 524 (1929).

[12b] Ellett, Nature **114**, 931 (1924).

[13] Schüler and Brück, Zeits. f. Physik **58**, 735 (1929).

thallium as well as cadmium[4] there is a difference of nuclear moments of the isotopes. The presence of an isotope having zero nuclear moment would result in higher values for the polarization, but the presence of such an isotope is not indicated by the spectroscopic data. An isotope with nuclear moment greater than one half probably would lead to even lower values of the expected polarization as a higher nuclear moment means an increased multiplicity of hyperfine structure levels. This might be offset by the changed intensity distribution in the exciting light, as it would be to some extent at least if the components of the hyperfine structure pattern responsible for the polarization should have the same wave-length in the patterns of both isotopes. Whether such a situation exists we cannot say without a more detailed spectroscopic analysis of the thallium hyperfine structures.

Sodium

For a sodium atom without nuclear moment resonance radiation excited by the D_2 line alone should be 60 percent polarized. When excited by D_1 and D_2 together, using a source such that the effective intensity ratio of the two lines is 2:1 (the theoretical value) the predicted polarization is 50 percent. In 1925 the writer found[14] 16.3 percent for the polarization of resonance radiation excited by both D lines, which corresponds to a value of 20.1 percent for D_2 alone not 24 percent as Datta states.[15] Datta, exciting by means D_2 alone got values as high as 33 percent with the resonating vapor in equilibrium with metallic sodium at 115°C. He supposed that the lower value obtained by the writer was due to higher vapor pressure. This however is not the explanation, for the vapor pressure was actually less than the lowest at which Datta made observations; 95°C and 7.4 10^{-8}mm. Hg[16] as against his values of 115°C and 4.5 10^{-7} mm Hg. More recently the experiment has been repeated with a somewhat more powerful source and vapor pressures as low as $1.9 \cdot 10^{-8}$ mm Hg (80°C). The polarization was again found to be 16.3 percent. The reason for the difference between this value and Datta's is not obvious to the writer.

Schüler[17] has observed hyperfine structure of the D lines, and as the hyperfine structure observed in caesium appears to be due to a nuclear moment, I have calculated the polarization to be expected in D line resonance radiation on the assumption of various nuclear moments. In every case the value is less than it is for the atom without nuclear moment. As the sodium hyperfine structure is on a very small scale it has been assumed that the intensity of the exciting light is uniform across the hyperfine structure pattern. This amounts to supposing that the separations in the pattern are small compared to the Doppler line breadth at the temperature of the source. The temperature of the glass walls of the source used by the writer was about 750°C and

[14] Ellett, Jour. Opt. Soc. Am. **10**, 427 (1925).

[15] Datta, Zeits. f. Physik, **37**, 625 (1926).

[16] Vapor pressures extrapolated from data of Rodebush and De Vries, Jour. Am. Chem. Soc. **47**, 2488 (1925).

[17] Schüler, Naturwiss. **16**, 512 (1928).

the emission lines have a corresponding breadth, as I have shown by experiments on polarization of resonance radiation in strong magnetic fields.[18]

The results of these calculations are summarized in Table II.

TABLE II.

i	0	1/2	1	obs.
D_2	60	40.54	20.5	33[14]
D_1	0	0	00	00
D_1+D_2	50	33.3	16.6	16.3[3]

In this connection it should be mentioned that observations of alternating intensities in sodium band spectra indicate a quite large nuclear moment,[18] probably greater than one, which probably would lead to even lower values for the polarization.

In conclusion it may be said the presence of hyperfine structure due to nuclear spin cannot be neglected in calculating the polarization of resonance radiation and radiation excited by electron impact. Taking account of such hyperfine structures in certain thallium and sodium lines leads to lower values of the predicted polarization and consequently better agreement between theory and experiment. Experiments on the polarization of resonance radiation may yield the data required for the solution of hyperfine structure problems in cases where direct spectroscopic analysis is impossible, as in the case of overlapping of certain components in the hyperfine structure patterns because of Doppler breadth of the lines.

[18] Ellett, Phys. Rev. **29**, 904 (1927).

[18] F. W. Loomis and R. S. Mulliken agree from inspection of Loomis spectrograms of Na_2 bands that i is probably 3/2 or greater. (Verbal statement to the writer). See also Loomis and Wood, Phys. Rev. **32**, 223 (1928).