NOVEL METHOD OF SPECTROSCOPY WITH APPLICATIONS TO PRECISION FINE STRUCTURE MEASUREMENTS

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A new spectroscopic method which promises to yield very precise measurements of some atomic fine structure intervals is described. This technique exploits interference phenomena that can occur in the resonance fluorescence of an atom in which two of the excited Zeeman substates cross.

The method will be described in reference to the apparatus utilized for the determination of the helium $2\,^3P_1 - 2\,^3P_2$ separation. See Fig. 1(a). A beam of helium light is projected through a helium discharge tube and detected by a PbS cell sensitive to the helium 1-micron line. The discharge tube contains ~0.2 mm of pure He and is situated between the poles of a Varian 12-in. magnet. The discharge is maintained by a high-voltage radio-frequency source and is rather weak for best results.

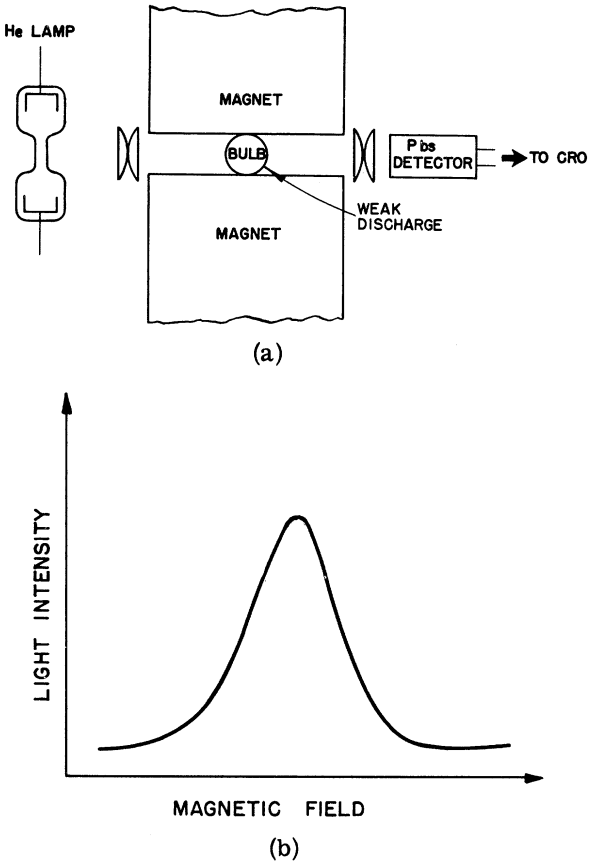

FIG. 1. (a) A schematic diagram of the apparatus employed in the helium experiment. (b) One of the $\Delta m = 2$ (600 gauss) lines as displayed on an oscilloscope.

With this apparatus a distinct increase in the light received by the detector is found at field strengths of 562.8 and 578.6 gauss. These signals can be displayed on the cathode-ray oscilloscope with a signal-to-noise ratio of ~100:1, and a line width of 2.7 gauss. See Fig. 1(b). The effects described can be observed with just the light from the discharge tube itself. However, the signal-to-noise is very poor because of the very low light intensity. Signals near 1300 gauss can also be observed if the incident light is appropriately polarized.

The explanation of these signals lies in the interference phenomena resulting from a crossing of two of the ^3P Zeeman sublevels. See Fig. 2. If the levels are all distinct (i.e., separations greater than their natural widths), then each contributes separately to the resonance scattering. However, when two levels are degenerate, their contributions to the scattering may interfere,

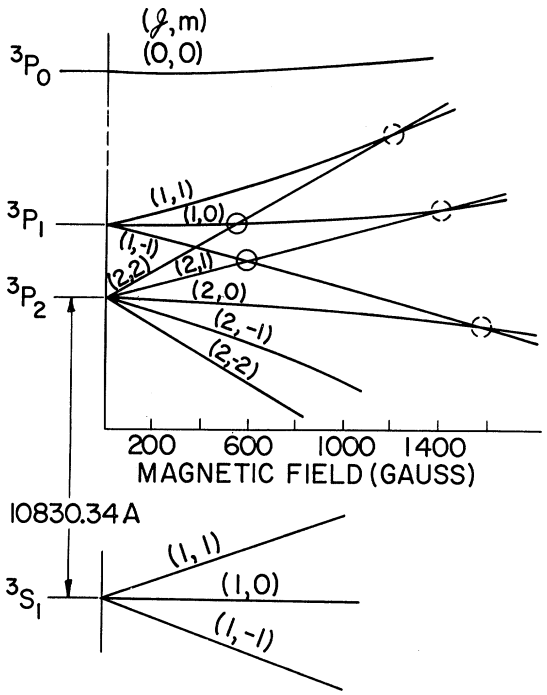

FIG. 2. A sketch of the helium energy levels pertinent to the $n = 2$ fine structure measurement.

thus giving rise to a change in scattered light.

In order to illustrate the calculation of this effect, let us consider a system having one ground state A and two excited states B and C. (A corresponds to one of the $^3S_1$ levels and B and C correspond to two of the ^3P levels.) To calculate the resonance fluorescence of this system, one needs first to calculate the rate at which photons of polarization $\vec{e}_1$ are absorbed and photons of polarization $\vec{e}_2$ are re-emitted. For the case where the levels B and C are well resolved (i.e., separated by more than the natural line width), one has

$$R \sim |(A|\vec{e}_2\cdot\vec{r}|B)(B|\vec{e}_1\cdot\vec{r}|A)|^2 + |(A|\vec{e}_2\cdot\vec{r}|C)(C|\vec{e}_1\cdot\vec{r}|A)|^2. \quad (1)$$

For the case where B and C have the same energy,

$$R \sim |[(A|\vec{e}_2\cdot\vec{r}|B)(B|\vec{e}_1\cdot\vec{r}|A) + (A|\vec{e}_2\cdot\vec{r}|C)(C|\vec{e}_1\cdot\vec{r}|A)]|^2. \quad (2)$$

It is clear that in order for Eqs. (1) and (2) to yield different results both terms in each expression must be nonvanishing; i.e., each of the levels B and C must be able to share the same photon. For the case of unpolarized light, one finds that the two degenerate levels must differ in m by 0 or 2 in order for there to be an interference term. With linear polarized light, one finds that crossovers of levels differing in m by 1 can be observed provided the direction of polarization is not parallel or perpendicular to the magnetic field. Levels differing in m by 3 or more will not interfere. By summing over the directions of $\vec{e}_2$, it can be shown that the total resonance scattering rate is not affected by the crossover of the two levels; only the angular distribution is changed.

This mechanism leads to a qualitative understanding of the observed signals. The two signals near 600 gauss are due to the crossing of levels with $\Delta m = 2$; signals near 1300 gauss are due to the crossing of levels with $\Delta m = 1$. No signal is observed near 400 gauss at the crossing of levels with $\Delta m = 3$. The observed line width is in approximate agreement with the calculated mean lifetime of the ^3P levels, and the intensity of the signal is in approximate agreement with that computed from the geometry of the apparatus and the relevant matrix elements.

The signal observed at 578.6 gauss arises from the crossover of the (2, 1) and (1, - 1) levels. The magnetic field at which this crossover occurs depends only upon the fine structure splitting $^3P_1 - {}^3P_2$ and well-known physical constants. By measuring the magnetic field, then, this splitting can be determined. The signal has been observed in several different bulbs varying in pressure from 0.16 to 1.5 mm and under different discharge conditions. There is no evidence, within our accuracy, of a pressure shift or a sensitivity to discharge conditions. We find for the splitting:

$$^3P_1 - {}^3P_2 = 2291.56 \pm 0.09 \text{ Mc/sec.}$$

The major contribution to the error interval arises from the inhomogeneity of the magnetic field over the discharge tubes which had volumes of several cubic inches. Experiments in progress will utilize smaller volumes. Our result is to be compared with the only other reported precision measurement, that of Wieder and Lamb[1]:

$$^3P_1 - {}^3P_2 = 2291.72 \pm 0.35 \text{ Mc/sec.}$$

The magnetic field at which the (2, 2) - (1, 0) crossover occurs depends slightly on the $^3P_1 - {}^3P_0$ separation. Using our determination of the $^3P_1 - {}^3P_2$ separation and the measured magnetic field of this crossover, we find

$$^3P_0 - {}^3P_1 = 29\,650 \pm 280 \text{ Mc/sec.}$$

The large error indicates the slightness of the field dependence of the crossover on this separation. This determination is to be compared with the only measurement of comparable accuracy, the optical determination of Brochard et al.[2]:

$$^3P_0 - {}^3P_1 = 29\,640 \pm 300 \text{ Mc/sec.}$$

It would be very desirable to measure the crossover of the (0, 0) and (2, 2) levels, which occurs in the neighborhood of 8300 gauss, because this would yield a very accurate measurement of the $^3P_0 - {}^3P_1$ separation. A preliminary search for the signal due to this crossover was made without success because the relatively simple discharge geometry is very susceptible to plasma oscillations at fields in excess of 3000 gauss. Work is in progress on discharge designs that should obviate this difficulty.

The method discussed in this Letter should be applicable to the measurement of the fine structure splitting $^2P_{3/2} - {}^2P_{1/2}$ in hydrogen, as well as other fine and hyperfine structure separations. Experiments along these lines are in progress.

A detailed analysis of the effects so briefly

described in this Letter is in preparation and will be submitted to the Physical Review.

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[1]I. Wieder and W. E. Lamb, Phys. Rev. 107, 125 (1957).

[2]Brochard et al., J. phys. radium 13, 433 (1952).

MAGNETIC DIPOLE RESONANCE OF EXCITED LEVELS IN FREE IONS OF Cd II AND Zn II

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In exciting by electron impact free Cd and Zn atoms in an atomic beam, one also obtains ionic excited levels. In particular, if an electron is pulled out of the d shell of the atomic ground state, $^1S_0\ d^{10}s^2$ ($3d^{10}4s^2$ and $4d^{10}5s^2$ for Zn and Cd, respectively), this leads directly to the ionic excited level, the inverted doublet, $^2D\ d^9s^2$. The only ionic levels of lower energy are the ionic ground state $^2S_{1/2}\ d^{10}s$, and the doublet $^2P\ d^{10}p$.

The observed transitions,[1] from the $^2D\ d^9s^2$ doublet to lower states, are the two-electron jumps $^2D \rightarrow {}^2P$:

	For Cd II	For Zn II
$^2D_{5/2} \rightarrow {}^2P_{3/2}$	4416 A	7479 A
$^2D_{3/2} \rightarrow {}^2P_{3/2}$	3536 A	6215 A
$^2D_{3/2} \rightarrow {}^2P_{1/2}$	3250 A	5894 A

We have observed the magnetic dipole resonance of the 4416A Cd II line and of the 5894A Zn II line by measuring polarization differences $I_\pi - I_\sigma$ as a function of the steady magnetic field. The experimental arrangement and method of observation were similar to the classical experiment of the Kastler-Brossel[2] type, except that in our case the excitation was by electron impact instead of an optical method.

We conclude that the $^2D\ d^9s^2$ doublet Zeeman sublevels become unequally occupied through this ionization process.

We have determined lifetimes of the $^2D_{5/2}$ state of Cd II and the $^2D_{3/2}$ state of Zn II by the method given in Brossel's paper,[2] and the polarization percentage of the 5894A line of Zn II.

The mean lifetimes are:

$^2D_{5/2}$ of Cd II: $T_e = (8.3 \pm 0.7) \times 10^{-7}$ sec,

$^2D_{3/2}$ of Zn II: $T_e = (4.65 \pm 0.2) \times 10^{-7}$ sec.

It is of interest to note that these lifetimes are relatively long compared to lifetimes of 10^{-8} - 10^{-9} sec of levels corresponding to allowed transitions. It is reasonable that a two-electron jump has a longer lifetime than a one-electron jump. Incidentally, we note that a one-electron jump with $\Delta l = 2$ (electric quadrupole transition) is 10^6 times less probable than a $\Delta l = 1$ transition (electric dipole transition), and therefore still 10^3 - 10^4 times less probable than the two-electron jump transition we observed.

Nevertheless, the two observed lines are still as strong as allowed atomic transitions. We assume that the reason for this high intensity is a large d-shell ionization cross section. Besides, to arrive at the $^2D\ d^9s^2$ state, the processes of ionization and excitation necessarily occur in one step.

Figure 1 shows the polarization percentage of the 5894A line of Zn II versus electron energy, compared to the excitation function; the polarization percentage being defined as $p = 100(I_\pi - I_\sigma)/(I_\pi + I_\sigma)$. We deduce for the optimum population ratio of the $^2D_{3/2}$ level Zeeman sublevels (corresponding to 8% polarization):

$$\text{population}\ (\tfrac{3}{2})/\text{population}\ (\tfrac{1}{2}) = 0.805.$$

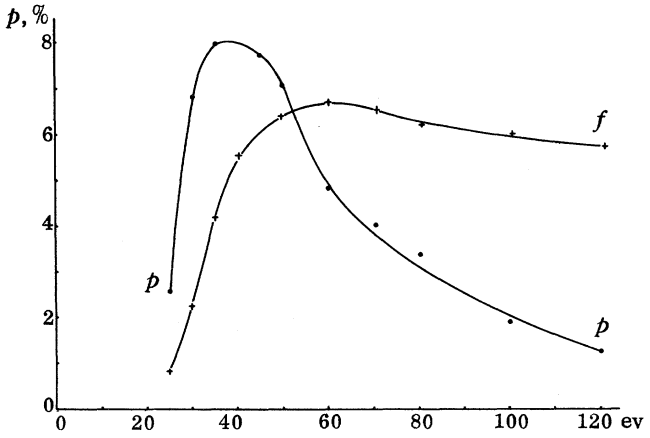

FIG. 1. Percentage polarization p in % and excitation function f in arbitrary units of the 5894A line of Zn II versus electron energy in electron volts.