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Spectroscopy of the Solid State: Potassium and Calcium*

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Utilizing the techniques developed by O'Bryan and Skinner for the study of the emission bands of metals, it has been possible to determine the structure of the conduction bands of the metals, potassium and calcium. The spectrograph used utilizes a photomultiplier in place of a photograph plate, thus enabling higher sensitivity and ease of operation. The particular radiation studied is produced by transitions of the conduction electrons into the ionized $3p$ state of the atom. The limiting factor in the accuracy of the emission band obtained in this way is the purity of the sample studied. The contamination of the surface of an evaporated sample affects the resultant curve mainly as a result of insufficient penetration of the electrons, since the photon absorption is negligible at depths required to produce radiation representative of the pure bulk metal. The results for potassium and calcium at temperatures between 25°C and 100°C are not in good agreement with previous theoretical calculations. The experimental values of band width are 1.9 and 3.0 ev, respectively, compared with the theoretical calculations of 3.6 and 5.0 ev.

I. INTRODUCTION

By means of spectrographic techniques originally developed by O'Bryan and Skinner[1] it has been possible to determine the energy distribution of the valence electrons in metallic potassium and calcium. If a sample of the metal is bombarded with electrons of several hundred volts energy it is possible to observe an emission band whose intensity distribution is directly related to the density of occupied states in the conduction band. The transition studied in this work is the valence band to $3p$ level, having a short wavelength limit of 500 and 670A for calcium and potassium, respectively. This wavelength radiation lies in the extreme ultraviolet and requires a spectrograph evacuated to pressures of 10^{-5} mm Hg or better, utilizing a grazing incidence concave grating. The instrument used, described elsewhere in the literature,[2] consists of two separate vacuum systems; one containing the spectrograph proper and the other containing a copper target, electron gun, and evaporating furnace for preparation of the sample. The two systems are separated by a sliding-type vacuum valve and the specimen chamber is maintained at a vacuum of 10^{-6} mm Hg to assure minimum contamination of the specimen. The spectrograph is of the same type as that used by O'Bryan and Skinner except that the photographic plate has been replaced by a Be-Cu photomultiplier which may be driven by an external coupling along the focusing trajectory of the spectrograph. This modification greatly enhances the sensitivity and flexibility of the apparatus.

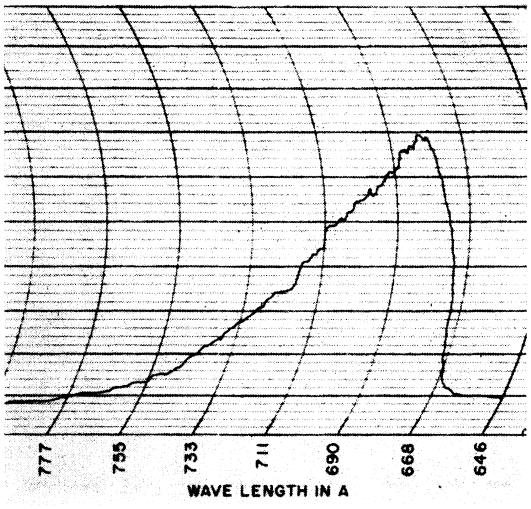

FIG. 1. Band emission curve of potassium. $E_b=300$ volts. $I_b=3$ ma. Counting rate=500 counts/sec full scale.

II. EXPERIMENTAL PROCEDURE

The potassium used for this work was reagent grade metal packed in kerosene. For evaporation purposes the metal was rinsed in toluene and cut in small pieces before being placed in the evaporating cup. In this way the active surface of the material was not exposed until the toluene had evaporated during evacuation of the chamber.

After the specimen chamber, containing the evaporating furnace, target, and electron gun, had been evacuated to a pressure of 10^{-6} mm Hg, the furnace and gun were turned on and the system allowed to outgas. When a suitable vacuum had again been obtained, the valve separating the specimen chamber from the spectrograph was opened and the target adjusted to obtain maximum photon counting rate.Then, with the detection system set at a wavelength corresponding to the maximum intensity of the emission band, the power to the evaporating furnace was increased and evaporation continued until the observed radiation reached a steady maximum in intensity. At

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[1] H. M. O'Bryan and H. W. B. Skinner, Phys. Rev. 45, 370 (1934).

[2] E. M. Gyorgy and R. H. Kingston, Phys. Rev. 83, 220(A) (1951); Piore, Harvey, Gyorgy and Kingston, "A high vacuum recording spectrograph for the study of radiation from solids in the 100–800A range" (to be published, Rev. Sci. Instr.).

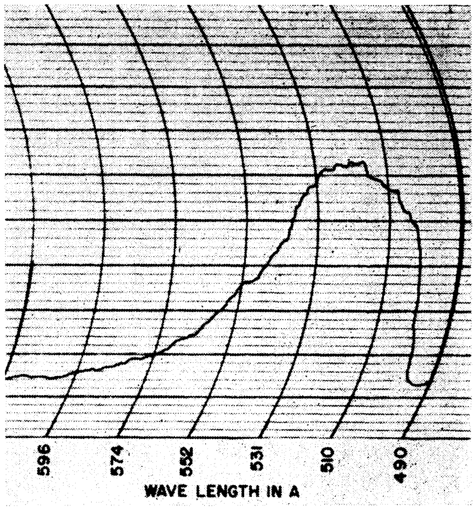

FIG. 2. Band emission curve of calcium. $E_b=500$ volts. $I_b=6$ ma. Counting rate $=500$ counts/sec full scale.

this point the photomultiplier was moved to a wavelength slightly shorter than that of the emission edge and a curve such as shown in Fig. 1 was recorded. A total of a dozen curves were taken, with several values of bombarding voltage and varying evaporation cycles; that shown was taken several minutes after the metal had been evaporated onto the target and the furnace shut off. It is representative of all the curves taken within a few minutes of the evaporation. An interesting point is that at voltages of 300 volts and higher the curves reproduce well; however, if the voltage is decreased to 150 volts, a sharp peak no longer appears at the high energy end of the band. The same results occur if the curve is taken ten or more minutes after the evaporation is stopped. This is interpreted as meaning that the penetration of the electrons is not sufficient at 150 volts to produce radiation representative of the pure bulk metal. In all probability the loss of the high energy peak is a result of oxide forming on the surface of the specimen. The fact that this effect is observed at higher voltages if the target has had time to deteriorate would indicate a slow penetration of the contaminating atoms into the interior of the metal.

Much more difficulty was experienced in obtaining the calcium emission band as a result of the contamination of the sample. There are two reasons for this. First, calcium does not melt before it reaches a temperature sufficient to evaporate metal onto the target; this means that an oxidized layer may easily form on the surface of the solid calcium, preventing the pure metal atoms from leaving the material. Second, calcium does not form a protective coat as do metals such as potassium and aluminum; instead of a thin layer of oxide forming on the surface and protecting the metal, the oxygen atoms seem to penetrate into the bulk of the metal in a matter of seconds. Because of this the curve shown in Fig. 2, one of approximately 30 curves, had to be taken with the evaporating furnace still running. The calcium on the target was thus being replenished continually with pure metal. To assume maximum purity the metal chips were first ground by means of a mortar and pestle to expose as much fresh surface as possible before being evaporated. Even with these precautions it was not possible to take more than one accurate curve for each filling of the evaporating cup. A second plot taken with the furnace still in operation produced a curve of the same general shape, but decreases in intensity by a factor of 50 to 75 percent. The behavior of the curve at the emission edge was checked carefully by starting the record at the peak of the edge, thus starting the curve only a few seconds from the time a satisfactory coat was on the target. In this way it was definitely established that the rising intensity with increasing wavelength was an accurate observation. If the evaporating furnace is shut off before the curve is started the result is a band emission curve which looks like a decreasing exponential, since the decrease in intensity as a function of time because of contamination completely masks the true shape of the emission band. This same effect may be observed if the photomultiplier is set at the peak of the emission band and the intensity of radiation as a function of time is studied at the one particular wavelength. The time required for the intensity to drop to one-half of its maximum for potassium. On the whole this seems to indicate that the behavior of metallic surfaces is not simply related to the activity of the free atom. These effects should certainly be considered in any future work on the band emission of solids.

A curve of a contaminated sample of calcium is shown in Fig. 3. Note how the intensity has fallen to almost half its value and the maximum has shifted to longer wavelengths. This particular curve was run ten

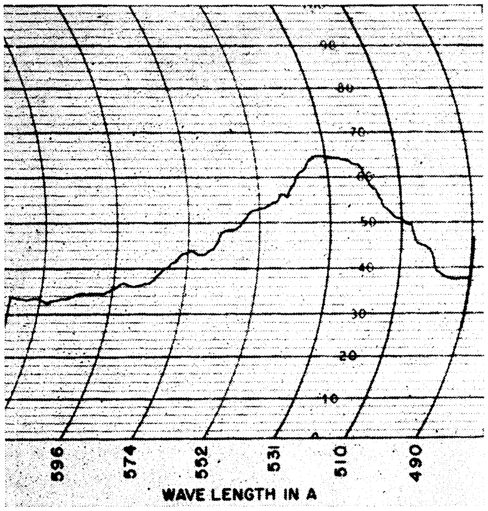

FIG. 3. Band emission curve of contaminated calcium. $E_b=330$ volts. $I_b=6$ ma. Counting rate $=200$ counts/sec full scale.

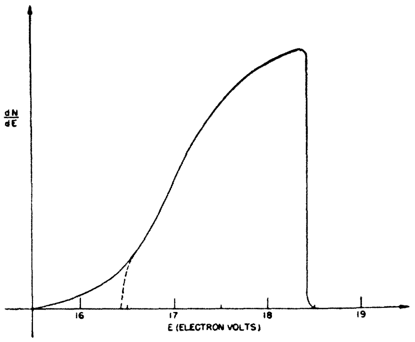

FIG. 4. *S* and *d* electron distribution in potassium.

minutes after the evaporating furnace had been turned on. Although the coat was being slowly replenished, oxidation of the metal in the cup and on the target caused a high percentage of impurities in the target. As was observed in potassium it was impossible to obtain any representative curve at all at 150 volts bombardment potential, even though the record was started at the beginning of the evaporation cycle.

During all the runs taken the object slit was set at 0.25-mm width and the image slit at 1.0 mm. This corresponds to a resolution of approximately 5A in the region studied. The calibration of the spectrograph was made at this same slit setting so that the wavelength of the emission edges should be accurate within 1 or 2A, if this value is measured at 50 percent of the maximum value of intensity at the emission edge. The calibrating edge was that of aluminum in the third and fourth order, corresponding to wavelengths of 510 and 680A.

The target temperature was somewhere between 25 and 100°C since during all the experiments the copper target was filled with tap water. Since the input power to the target was of the order of 1.5 watts there was not enough heat to cause boiling so that an accurate value of temperature was not obtained. It is important to realize that the temperature of the body of the target does not correspond to the temperature of the active surface. Actually the metal at the surface could theoretically be hot enough to melt. The only investigation of this problem has been made by Skinner,[3] who made an accurate study of the width of the emission edge as a function of target temperature. By relating this width to the Fermi distribution theoretical width he concluded that the temperature of the active surface was close to the over-all temperature. Since Skinner's input power to the target was much higher than ours it is reasonable to assume that the temperature of the sample in our work was within a few degrees of the target temperature.

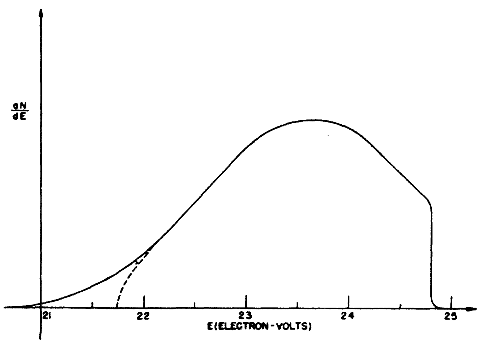

FIG. 5. *S* and *d* electron distribution in calcium.

In the work described the bombardment potential was kept as low as possible to eliminate the high background which might be produced by excitation of higher energy states, specifically those in the 2*s* and 2*p* shells of the atom. As has been observed by other workers the conduction band-to-3*s* transition is not observed in these metals because of the high probability of an intermediate 3*p*-to-3*s* transition.

III. INTERPRETATION OF DATA

The experimental curves may be modified to represent the density, dN/dE, of the *s*-type and *d*-type electrons according to a method given by Skinner.[3] The background is first subtracted from the emission curve and then the resultant distribution replotted as a function of quantum energy; the ordinate having been divided by a factor proportional to the fourth power of the energy. The final curve then represents the density of electrons weighted according to their particular wave function. For example, if the wave function of electrons at some specific point in the band is composed of *s* and *p* functions, the resultant plot will give only the contribution of the *s* part of the function since the final state in the transition is a *p* state. For simplicity the emission edge has been sharpened to correspond to the shape of the theoretical edge obtained from the Fermi statistics at the temperature used. This is not a strictly accurate interpretation since it cannot be said definitely that the edge would be this sharp if the spectrograph were operated at its limiting resolution. The width of the edge, however, as measured on the experimental curves corresponds to the calculated resolution of the spectrograph and it is therefore assumed that the above alteration is reasonable. The energy at the emission edge was determined by a direct comparison with higher orders of the aluminum edge obtained by Skinner. The cross calibration was made at the midpoint of the experimental edge.

Since potassium is a monovalent metal, it is quite well described by the Sommerfeld free-electron approximation. Figure 4 shows the distribution of electrons as a function of energy as determined from the experimental curves. The dotted line at the low energy end of the curve shows the probable position of the bottom of the Brillouin zone. The low energy "tail" on the curve is a common characteristic of emission bands and

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[3] H. W. B. Skinner, Trans. Roy. Soc. (London) **A239**, 95 (1940).

has been discussed by Skinner[3] and Seitz.[4] There are several explanations for the effect. Skinner concludes that it is a result of Auger transitions during the radiative process while Seitz attributes it to either contamination or the vestiges of excited states formed below the band. A detailed analysis applied to sodium is given by Landsberg.[5] In this curve and the curve for calcium a parabola has been fitted to the bottom of the band, since it is known that electrons in this region behave according to the free electron theory. The position of the estimated limit of the band cannot be determined accurately because of this phenomenon, but a reasonable value of the band width is 1.9±0.2 electron-volts. It is of interest to note that the Sommerfeld approximation gives 2.1 volts for this value, while a Wigner-Seitz calculation of the effective mass of the electrons at the bottom of the zone gives a band width of 3.6 volts.[6] Evidently the wave functions of the electrons in the potassium band are predominantly of the *s*-type, since the over-all curve is a good representation of the parabolic distribution which would be expected in this case.

The calcium conduction band shows a shape much different than the free electron theory would predict. Figure 5 indicates that the first Brillouin zone is nearly filled, since the density of states falls off well before the high energy edge. The second zone must necessarily overlap with the first since the metal is a conductor; however there is an uncertainty as to the wave functions at the bottom of this excited band. There is certainly no definite indication of an overlapping curve such as may be seen in Fig. 6, taken from data obtained for aluminum.[2] Even though calcium has two valence electrons, compared with three for aluminum, Skinner's curves[3] for magnesium indicate that even in the divalent case an overlap should be observed. There are two possible explanations for the lack of structure in the calcium case. First, it is possible that the wave functions at the bottom of the second band are *p*-type, therefore not observable in a transition to the $3p$ state; or second, the excited band rises so slowly that it would not produce any observable inflection in the curve. Manning and Krutter's calculations[7] indicate a *d* band as the overlapping zone; however, examination of their predicted curve in Fig. 7 shows that the calculations cannot be too sound since the band widths are in very poor agreement. The observed band width is 3.0±0.2 volts, Manning and Krutter's value, 5.0 volts and the free-electron determination, 4.3 volts. It is not possible to determine from the above authors' calculations whether the electrons in the bottom of the *d* band are *s*-, *p*-, or *d*-type. If the band is as sharp as their calculations would indicate then *p*-type functions would be expected, since a sharp rise is not visible in the experimental curve; however, there is possibly an overlap into the $4p$-derived band rather than the $3d$, in which case it would most likely be fairly broad and would therefore not be obvious in the curves. The peak of the density curve does not necessarily correspond to the maximum number of electrons per unit energy range since there is every indication that the electron wave function changes from *s* to either *p* or *d* as the eigenvalue increases in magnitude. It may certainly be stated, however, that the degeneracy at the top of the band is as high if not higher than the observed curve indicates, contrary to the theory. A survey of the literature indicates that there are no data on the Hall coefficient for calcium. When such measurements are made, it should prove instructive to ascertain the relation of this coefficient to the filling of the first Brillouin zone. Since this zone is almost completely full it is highly probable that the coefficient would have the anomalous sign, that is, the negative mass effect in the quantum mechanical treatment would cause the current carriers to appear as positive charges.

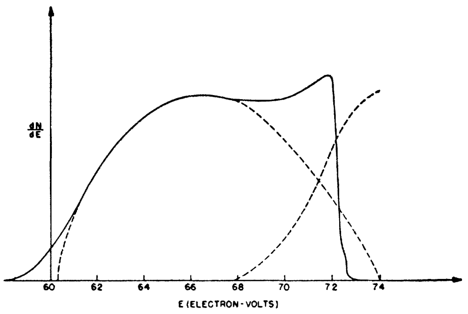

FIG. 6. *S* and *d* electron distribution in aluminum. (Dashed lines indicate probable zone structure.)

The experimental results for potassium and calcium are summarized in Table I.

IV. GENERAL DISCUSSION OF METALLIC ENERGY BANDS

Table II is a survey of the theoretical and experimental results obtained for the electronic structure of

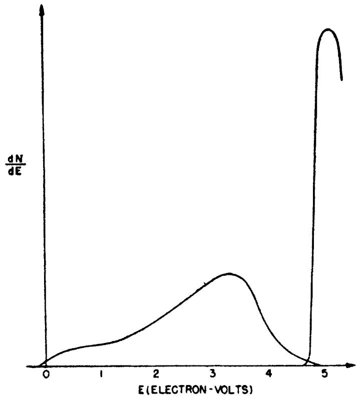

FIG. 7. Manning and Krutter's calculated distribution curve.

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[4] F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), p. 439.

[5] P. T. Landsberg, Proc. Phys. Soc. (London) **A62**, 806 (1949).

[6] See reference 4, p. 354.

[7] M. F. Manning, H. M. Krutter, Phys. Rev. **51**, 761 (1937).

TABLE I. Summary of experimental results for potassium and calcium.

	Band width (ev)	Emission edge	
		Wavelength (A)	Energy (ev)
Potassium	1.9±0.2	675±2	18.4±0.1
Calcium	3.0±0.2	500±2	24.8±0.1

metals from lithium to zinc in the periodic table. The values of the observed band width for titanium through zinc, taken from an unpublished communication from Skinner, are not considered too reliable, since the emission edges are not at all sharp. The free-electron band widths for the transition metals have not been calculated, as the value obtained would be in complete disagreement with the actual band widths, since the *d* electrons are known to be tightly bound. The cellular calculations for the transition metals are also unreliable, since they are only qualitative values obtained by Slater by extrapolating Krutter's results for copper. The lithium value has been recalculated by Silverman[8, 9] giving a value of effective mass in closer correspondence with the observed band width.

The calculations show generally good agreement for the first two periods of the table; however, the potassium and calcium band widths are closer to the free electron value than that of the Wigner-Seitz calculation, and the work on the transition metals is too sketchy to make any generalizations. It is interesting that the Fletcher and Wohlfarth calculation for nickel gives a band width just one-half the value of that obtained by Slater using the cellular method. If similar results were to be expected for potassium and calcium using the Bloch perturbation method the band widths would correspond much more closely with the experimental values. It is not immediately obvious why the Bloch calculation gives a band width corresponding to much tighter binding than the cellular method. It may be that the presence of a 3rd level with energy very close to the 4*s* energy, and yet a mean radius corresponding to that of the 3*p* level, introduces complications not taken into full account in the cellular method. This might be a result of the fact that the Bloch scheme seems to take a much more accurate account of exchange interactions. Slater[10] has proposed an intermediate method of calculating the interaction potentials and a trial calculation is now being carried out by Parmenter[11] to determine the validity of this approach. The idea is to modify the Hartree-Fock equations in such a way that the potential seen by the electron is somewhere between the extreme of the Bloch method, where neighboring cells are assumed to contain no valence electrons, and the cellular method, where the neighboring cells are treated as electrically neutral.

TABLE II.* Summary of band emission data and theoretical calculations. (All energies given in electron-volts).

Metal	No. of valence electrons	Band width			
		Free electron	Bloch pert.	Cellular	Obs.
Li	1	4.7	···	3.1[b]	3.7±0.5
Be	2	14.3	···	11.8[c]	13.8±1.0
Na	1	3.2	···	3.2[d]	2.5±0.3
Mg	2	7.2	···	···	6.2±0.3
Al	3	11.7	13.0	···	11.8±0.5
K	1	2.1	···	3.6[b]	1.9±0.2
Ca	2	4.3	···	5.0[e]	3.0±0.2
Ti	4	···	···	1.8[f]	5.6±0.5
Va	5	···	···	2.5[f]	5.6±0.5
Cr	6	···	···	3.1[f]	6.3±0.5
Mn	7	···	···	3.7[f]	6.0±0.5
Fe	8	···	···	4.3[f]	4.4±0.5
Co	9	···	···	4.6[f]	5.8±0.5
Ni	10	···	2.4[a]	4.9[f]	4.7±0.5
Cu	1(?)	7.1	···	7.3[g]	7.0±0.5
Zn	2(?)	9.5	···	···	11.0±0.5

* Note: Observed band widths are taken from reference 3 for Li to Al; from this paper, for K and Ca; and the remainder from Skinner's data mentioned in the text.

[a] G. C. Fletcher and E. P. Wohlfarth, Phil. Mag. **42**, 106 (1951).

[b] Based on calculation of the effective mass; see reference 4, p. 354.

[c] C. Herring and A. G. Hill, Phys. Rev. **58**, 132 (1940). Not strictly a cellular method, but included for reference.

[d] J. C. Slater, Phys. Rev. **45**, 794 (1934).

[e] M. F. Manning and H. M. Krutter, Phys. Rev. **51**, 761 (1937).

[f] J. C. Slater, Phys. Rev. **49**, 537 (1936). Values extrapolated from Krutter's work on copper.

[g] H. M. Krutter, Phys. Rev. **48**, 664 (1935). See also S. R. Tibbs, Proc. Cambridge Phil. Soc. **34**, I, 89 (1938).

V. SUMMARY

The experimental work described has definitely established the fact that the emission edges of the metals potassium and calcium are sharp as has been observed in aluminum and the preceding conductors in the periodic table. On the basis of these results it seems possible that future work on the transition metals, with the more sensitive techniques described, will produce better resolution than that obtained by Skinner, above. A study of the absorption coefficients of metals for radiation of the wavelengths[12] used has shown that the important factor in determining the reliability of the observed band emission curves is the depth of penetration of the bombarding electrons. For the metals described in this paper the necessary bombarding potential has been found to be 300 volts or higher. There is of course the possibility that the metals which have *d* band structure may have higher electron absorption, in which case the voltage should be raised to take this into account. A thorough study of electron penetrations at voltages of 100 to 1000 volts would be an extremely valuable addition to the literature, as well as further studies on photon absorption in the 100 to 800A range.

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[8] R. A. Silverman and W. Kohn, Phys. Rev. **80**, 912 (1950).

[9] R. A. Silverman, Ph.D. thesis, Dept. of Physics, Harvard University (1951).

[10] J. C. Slater, Phys. Rev. **81**, 385 (1951).

[11] R. H. Parmenter, Quarterly Profress Report, Solid State and Molecular Theory Group, M.I.T., Cambridge, Massachusetts (July 15, 1951), p. 15.

[12] H. W. B. Skinner and J. E. Johnston, Proc. Roy. Soc. (London) **A161**, 420 (1937); J. E. Johnston, Proc. Cambridge Phil. Soc. **35**, 108 (1939).

Another important result of the work is the observation that the chemical activity of a metal is not a decisive factory in determining its resistance to oxide contamination. Before intensive studies of the transition metals are taken up it seems advisable that the protective layer effect as observed in potassium should be investigated more thoroughly, probably by a survey of the metallurgical literature.

The use of the photomultiplier in place of photographic plates has added another variable to the experimental observations. This is the possibility of studying time-dependence of the radiation from the sample. The high sensitivity and the foregoing effect suggest possible studies of surface effects in metals and nonconductors.

ACKNOWLEDGMENT

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A Phenomenological Theory of the Lamb Shift and of Anomalous Magnetic Moments

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If one assumes that self-interactions don't exist and can be ignored, the explanation of the anomalous magnetic moments of proton and electron and the Lamb shift require new types of interaction between protons, photons, and electrons. A theory with such additional interactions is here developed in gauge-independent form, thus avoiding longitudinal and scalar photons throughout. The covariance of the formalism is proved. The resulting modification of Maxwell's equations for charged elementary particles in the vacuum involves only the introduction of an intrinsic polarization and magnetization of elementary particles, so that these equations for the vacuum now take the familiar form of the macroscopic Maxwell equations in matter. A further modification of the Dirac equation for electrons and protons involves the introduction of a delta-function interaction between them, opposite in sign to the Coulomb interaction. It is indicated how one can derive an energy density tensor for this theory. It is then shown how this formalism explains the anomalous moments and the Lamb shift. The three new interaction constants are adjusted to the experimental data. The dependence of the Lamb shift on the quantum numbers automatically comes out to be nearly the same and the dependence on the atomic number to be exactly the same as in the theory of Bethe, French and Weisskopf.

1. INTRODUCTION

THE Lamb shift[1,2] and the anomalous magnetic moment of the electron[3,4,4a] have been explained as consequences of self-interaction of the electron through the radiation field.[5–8a] The necessity of infinite "renormalizations" or "subtractions" or of other special devices to make divergent expressions finite makes this theory not fully satisfactory. There have also been attempts at explaining the anomalous magnetic moment of nucleons[9–11] as a consequence of their mesic ("pionic") self-interaction.[12] These theories too are not quite satisfactory.

Instead of attempting to improve these existing theories, we want to attack these problems in this paper from an entirely different point of view. It has been suggested by this author[13] that perhaps no self-interactions exist at all in nature. Although this hypothesis formulated in this oversimplified form is subject to various serious objections, which the author intends to discuss in a forthcoming paper, and which need further investigations for their complete solution, it might yet be interesting to examine already now, how

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[1] W. E. Lamb and R. C. Retherford, Phys. Rev. **72**, 241 (1947).

[2] R. C. Retherford and W. E. Lamb, Phys. Rev. **75**, 1325 (1949); W. E. Lamb and R. C. Retherford, Phys. Rev. **79**, 549 (1950); and **81**, 222 (1951).

[3] P. Kusch and H. M. Foley, Phys. Rev. **72**, 1256 (1947); H. M. Foley and P. Kusch, Phys. Rev. **73**, 412 (1948).

[4] P. Kusch and H. M. Foley, Phys. Rev. **74**, 250 (1948).

[4a] S. Koenig, A. G. Prodell, and P. Kusch, Phys. Rev. **83**, 687(L) (1951).

[5] H. A. Bethe, Phys. Rev. **72**, 339 (1947).

[6] J. B. French and V. F. Weisskopf, Phys. Rev. **75**, 1240 (1949).

[7] Bethe, Brown, and Stehn, Phys. Rev. **77**, 370 (1950).

[8] Similar results were obtained by J. Schwinger and others using manifestly covariant methods, in which, however, the non-normalizability of the state vector makes the derivations somewhat more doubtful.

[8a] R. Karplus and N. Kroll, Phys. Rev. **77**, 536 (1950).

[9] H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949).

[10] Sommer, Thomas, and Hipple, Phys. Rev. **80**, 487 (1950).

[11] C. D. Jeffries, Phys. Rev. **81**, 1040 (1951).

[12] See for example, S. Borowitz and W. Kohn, Phys. Rev. **76**, 818 (1949).

[13] F. J. Belinfante, Phys. Rev. **82**, 767 (1951); Progress of Theor. Physics **6**, 202 (1951).

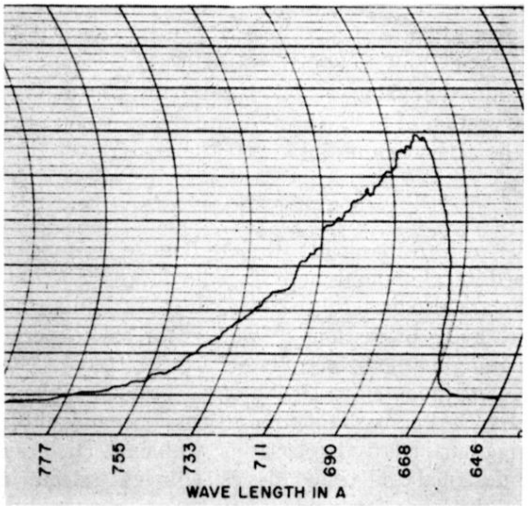


FIG. 1. Band emission curve of potassium. $E_b = 300$ volts. $I_b = 3$ ma. Counting rate = 500 counts/sec full scale.

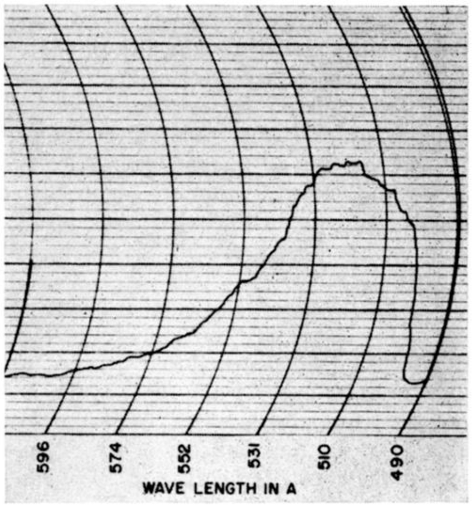
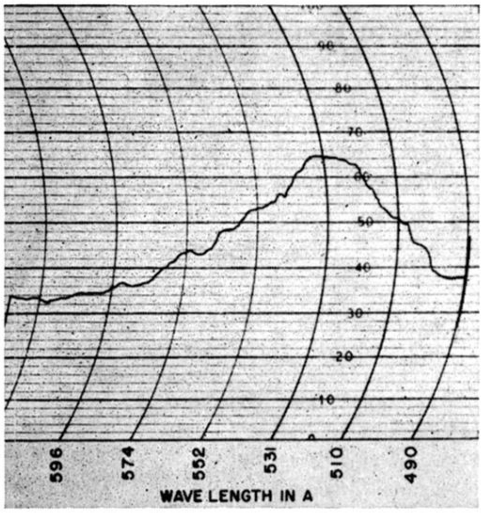


FIG. 2. Band emission curve of calcium. $E_b = 500$ volts. $I_b = 6$ ma. Counting rate $= 500$ counts/sec full scale.



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