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CONTENTS

The American Association for the Advancement of Science:

Scientific Events:

Discussion and Correspondence:

Quotations:

Scientific Books:

Special Articles:

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SOME RECENT DEVELOPMENTS IN THE STUDY OF THE SOLID STATE[1]

Since there are only three states of matter, one might say, just as a first guess, that the phrase "properties of solids" implied one third of all the knowledge of material things which has resulted from physical and chemical study. It is obvious, then, that only a few points can be touched upon in this discussion, and I must ask you to extend to me the modern privilege of applying my own "principle of selection" in any way I see fit. Even then, I am afraid it will be much like an extract from the well-known work of Mr. Wingleman, or an author's reading by Messrs. Landolt and Börnstein.

Without a doubt, the greatest advance of the past decade in the study of solids has been the application of the various X-ray methods of crystal analysis. This is particularly true in the case of the metals, and in other cases where the minuteness of the structure made the older methods difficult of application. The unique feature of the X-ray method, however, is, as Sir William Bragg has emphasized, that it gives a measure of the size of the crystal unit, out of which the crystal mass is built up. This unit is apparently much more characteristic and invariable than the external form of the crystal. Just how far the method can go in distinguishing slight differences between units can not yet be said, and as a consequence there is the possibility of larger repeating blocks built up of the presently accepted units, which may differ among themselves in ways at present indistinguishable. In this connection, there has been much discussion as to the status of the molecule in crystals. In some of the first crystals studied, rock salt and sylvine, for example, X-ray analysis gives no evidence of the existence of a "molecule"; in quartz, on the other hand, the molecule is easily distinguished,[2] and in organic crystals there is always a definite "crystal molecule" which will probably turn out to be identical with the chemical molecule. This question of the existence or importance of the molecule in crystals is just another way of putting the question of the nature and symmetry of attachments between atoms in the lattice structure, and other methods of study will un-

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[1] Address delivered as President of the American Physical Society, in Cincinnati, December 27, 1923.

[2] McKeehan, *P. R.*, No. 2, 1923, p. 206.

doubtedly help to solve the problem—such as, for example, the correlation of certain lines or bands of absorption or reflection with certain groups of atoms occurring in various compounds. The recent work of Schaefer and Schuberth[3] shows some striking examples of this sort of persistence.

The most interesting recent generalization dealing with crystal analysis is that of Shearer[4] connecting crystal symmetry with molecular symmetry. To each of the 32 crystal classes he assigns a symmetry number, which is simply the smallest number of perfectly asymmetrical molecules which can be arranged in a crystal unit so as to give to the unit cell the symmetry of each class. For example, the triclinic asymmetric requires but one molecule per cell, while the several classes of cubic crystals require 12, 24 or 48. Assuming, then, that actual crystal units are made up of the minimum possible number of molecules, and using the number per unit as determined from X-ray measurements of the size of the unit, combined with the density and the weight of the chemical molecule, it follows that if the actual number of molecules per cell is less than the symmetry number of the crystal in question, the molecules themselves must possess a definite amount of symmetry. This he expresses by putting the ratio

$$\frac{\text{Symmetry number of crystal}}{\text{Number of molecules per cell}} = \text{Symmetry number of molecule.}$$

In no case is the observed number of molecules per cell greater than the corresponding symmetry number of the crystal, and the generalization promises to be of great value in deciding as to the symmetry of the molecule as it exists in the crystal. Whether it retains the same degree of symmetry in the liquid and vapor state can not yet be said. Applying this idea to special cases, Shearer decides in favor of Hull's suggested structure (four interpenetrating hexagonal lattices) for graphite, rather than Debye's, and finds that Bragg's suggestion of a "puckered hexagon" for the benzene molecule is consistent with the crystal symmetry of benzene.

Crystal analysis may throw light not only on the question of molecular symmetry but also on that of atomic and molecular size, though until we know more about atomic structure and shape the term size can not mean much. Wyckoff[5] has discussed a mass of experimental data from this point of view, and concludes that the simple hypothesis of a constant atomic radius for each element in all crystals is inconsistent with existing observations, but that in isomorphous crystals composed of only two kinds of atoms, the center to center distances are additive, in accordance with this hypothesis.

Under the stimulus of the experimental study of crystal structure there has been an energetic and fruitful revival and extension of the old problem of the dynamics of point lattices, and at the hands of Madelung, Debye and especially Born, there has developed a very complete theory of the properties of crystals. According to Born, a crystal is considered as a system of interpenetrating point lattices, the corners of each lattice being occupied either by an electron or a positive nucleus, neither the molecule nor the atom being handled as a unit.

The potential of the system is taken as a minimum in the undisturbed state, and the forces acting in any disturbed state are derived from the displacements of all the particles from their equilibrium positions, through a potential function which for many purposes may be of a general quadratic form, although third and fourth power terms have been added in some cases. From this Born derives a theory of elasticity of the correct 21 constant type, and a simple theory of piezo-electricity. The expression for the specific heat is a combination of three terms of the Debye type

$$\int_0^{\frac{\theta}{T}} \frac{x^3}{e^x - 1}\,dx,$$

whose only constants (maximum frequencies) appear in the limits of integration, and, in general, a number of terms of the Einstein form

$$\frac{\frac{\theta_j}{T}}{e^{\frac{\theta_j}{T}} - 1}$$

each involving a definite "natural frequency" which determines ϑ_j; all these frequencies being expressible in terms of the elastic constants of the crystal. For a number of crystals, the natural frequencies computed elastically agree quite closely with those computed from infra-red reflection maxima, or residual rays. Furthermore, the general lattice theory leads to a dispersion formula of the Drude-Lorentz type in which the same natural frequencies may be used, with the usual degree of success. The general lattice theory, then, combines the results of the earlier dispersion theory and those of Einstein, Madelung and Debye in connecting dispersion with natural infrared frequencies, and these with the elastic properties and the specific heat. It is evident that the lattice theory will in general be of unique importance in connection with those characteristics for which the regularity of structure is directly responsible, such as

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[3] Schaefer and Schuberth, *Z. f. Phys.*, 7, p. 197, *et seq.*

[4] Shearer, *Proc. R. S. Lond.*, Feb., 1923.

[5] Wyckoff, *P. N. A. Sc.*, No. 2, 1923.

certain cases of optical rotatory power, and the non-isotropic optical properties of crystals, and it is an interesting question as to just what characteristics are of this nature. The lattice theory is so general that it may include the type of oscillator characteristic of an atom or group of atoms, which was the basis of the Drude-Lorentz theory of dispersion and it is perhaps only a matter of habit that the older manner of handling this case seems simpler and more vivid. The question as to the relative importance, in connection with any particular property, of the couplings inside the atom or inside a persistent group of atoms, and the inter-atomic couplings which are characteristic of the regular structure, is an experimental one quite aside from the theory. It is usually considered, for example, that the absence of the typical infra-red reflection maxima in solutions of rock salt, sylvine, etc., proves that these are produced by the crystal structure, but this may not always be justified, since the presence of the atoms of the solvent will have an influence quite apart from and perhaps exceeding that due to the loss of regular structure and possibly masking the effect of the still existing atomic resonators. If the idea of resonators is ultimately found to be the best basis for the dispersion theory, as it has so far proved to be, the indications are that there must be resonators of both the atomic and structural type, but the physical significance of Born's "natural frequencies," except in the special case where they are due to atomic resonators, is not easy to see. The same characteristic reflection (and dispersion) frequencies of quartz appear both in the crystalline and amorphous form,[6] though they show interesting modifications. In the crystalline forms of SiO_2 the maxima are quite complex, indicating a group of rather sharp resonance frequencies, while in the amorphous forms the fine structure is blurred out, as if the surroundings of the SiO_2 groups were less regular and the resonance therefore more variable. In contrast to this, the regular crystals of sodium chlorate ($NaClO_3$) and sodium bromate ($NaBrO_3$) show a rotatory power which is lost by solution or fusion, and which is therefore entirely structural in origin. Assuming for these substances one of the several detailed crystal structures which have been suggested as a result of X-ray analysis, Herman,[7] on the basis of the Born theory, arrives at a quantitative determination of the rotatory dispersion curve which agrees particularly well with experimental values in the case of sodium bromate. Of course, there are parameters available whose values may be chosen within limits—the point is that a reasonable choice of these parameters gives a satisfactory agreement.

In discussing high temperature phenomena one might expect that the terms in the potential function involving higher powers of the displacements would become important, and it is indeed in the computation of specific heat under such circumstances that Born and Brody[8] have included such terms. The result, holding only at high temperatures, shows a linear variation of specific heat with temperature, $C_v = 3R(1 - 3\sigma RT)$ where σ may be either positive or negative, but is probably negative for substances showing a high coefficient of expansion. Only for platinum and copper[9] are there data available for testing this conclusion, but for both of these and especially for platinum, for which more accurate data are available, the agreement is quite satisfactory.

The breaking strength of crystals has for many years been known to be quite definite and characteristic, but the attempt to compute it for an ionic lattice structure was unsuccessful, giving results many hundred times too large. At Debye's suggestion, account has been taken[10] of a thermal motion of the average amplitude of one sixth of the interionic distance, which leads to a very considerable reduction in the computed breaking strength. In this case, the potential function must be given an additional term to take account of the repulsive forces, and from this extended form may also be computed the lattice energy, which may be compared to the value computed from the heat of formation of the salt, the heat of sublimation of the metal, the ionizing potential of the metal, the heat of dissociation, and the "electron affinity" of the electro-negative atom. The agreement is qualitatively good in the case of NaI and KI, if the best spectroscopic value of the electron affinity of iodine is used.[11]

Of all the fields in which the lattice theory has been applied, least satisfactory progress has been made in that of heat conduction. In general, in dealing with vibratory motions, no dissipative term has been used, since here, as also in the corresponding classical case of linear oscillators, there is no satisfactory theory for the origin of a damping factor. Recent work of Cario and Franck has, in the case of vapors, given us definite proof of the transfer by impact of radiant energy absorbed by one atom to another, and this idea may be useful in several ways if it proves to be transferable to the case of solids. From this angle the "dissipation" of radiant energy in a solid would involve first the production of an excited state in one atom and then the transfer (by

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[6] Rubens, *Berl. Ber.*, 1919; Schaefer and Schuberth, *Z. f. Phys.*, 7, p. 197.

[7] Herman, *Z. f. Phys.*, 16, p. 103, 1923.

[8] Born and Brody, *Z. f. Phys.*, p. 132, 1921.

[9] Magnus, *Z. f. Phys.*, 7, p. 141, 1923.

[10] Zwicky, *Phys. Z.*, 24, p. 131, 1923.

[11] Born and Gerlach, *Z. f. Phys.*, 5, p. 435, 1921.

impact) of this energy to the form of mass or thermal motion of another atom.

The classical theory of dispersion stands now in a peculiar position—having basic ideas quite foreign to the quantum theory, and yet having been practically unchallenged by that theory. Especial interest therefore attaches to Darwin's[12] preliminary account of an application of quantum ideas to dispersion. Darwin retains the classical ideas of the propagation of electro-magnetic waves in free space, but for the ordinary picture of the reaction of such waves with atoms he substitutes a sort of probability trigger action. If a polarized wave encounters an atom, there is a chance given by

$$A_n \left(\frac{\vartheta E_x}{\vartheta t} \right) dt$$

that it will, in the time dt, excite the atom to emit a spherical wave of the same type emitted by a Hertzian doublet and of the form $\alpha_n e^{-\lambda_n t} \cos k_n t$. The probability factor A_n, amplitude factor α_n and damping factor λ_n and frequencies k_n depend only on the atoms and not at all on the wave. As indicated by the subscript n there may be several different ways in which the atom may be excited. Conservation of energy is given up as applied to individual excitations, but maintained as an average. The effect at any point of a wave passing over a group of atoms is found by integration, and in this process because of the particular form assumed for the individual scattered waves, the frequencies of the atoms $(k_n/2\pi)$ disappear and the scattered wave is of the incident frequency. The resulting formula is shown to lead to a Lorentz type of dispersion formula

$$\frac{3(\mu^2 - 1)}{\mu^2 + 2} = \Sigma_n \frac{4\pi N A_n \alpha_n c^2}{k_n^2 - p^2}$$

Since the chance of excitation is assumed to depend linearly on the exciting force, the effects of superposed waves will on the average be additive, which leads to the conclusion that the atoms will act as Fourier analyzers, breaking up a pulse into its constituent simple harmonic wave trains.

We come now to another related and complex group of phenomena in which the influence of structure is undoubtedly important, but at present little understood—namely, phosphorescence, fluorescence, photoconductivity and the effect of light on the dielectric constant. In general, all these phenomena are in qualitative harmony with Lenard's basic idea that the effect of the exciting light is in all cases to move an electron from a normal to an "excited" position, supposedly in another atomic group. The return of the electron to the normal position after more or less delay may produce light, phosphorescent or fluorescent, according to the extent of the delay. The rival theory of Wiedeman was originally stated before the advent of the electron, but both of these and the theory of Kowalski are so general and embody such similar features that at present it is hardly possible to discriminate between them. Even Perrin's so-called chemical theory involves a molecular "change" produced by the exciting light, the emission occurring as a result of and during this change, which according to Perrin always results in the destruction of the active molecule. The field we are discussing having been exhaustively reviewed in the recent National Research Council Bulletin No. 30, I have only a few still more recent papers to consider.

Schmidt[13] has studied further the effect of an electric field upon the decay of phosphorescence of zinc sulphide (ZnS). Not only does the application of a field during decay produce a brief but considerable increase in luminescence, but also the removal of a field which has been on during the excitation and part of the decay period gives an even brighter flash. On the other hand, the process of excitation and decay goes on in just the same way in a steady field as with no field. A field which produces a flash when applied during the decay period produces no effect when removed, but several reapplications of it after short intervals will in general give repeated flashes. The flashes are brighter the stronger the field. Schmidt suggests an explanation, involving in the first case the rotation of polarized molecules by the field thus facilitating the return of excited electrons to their normal positions, and in the second case the holding of electrons in the excited position by the field, to be suddenly released upon removing the field, but this does not help much in correlating the phenomena. Phosphorescent zinc sulphide also shows[14] a marked increase in dielectric constant when exposed to violet or ultra-violet light, and this effect is decreased by the application of an auxiliary field,[15] though there seems to be a fairly definite minimum value of 1300 v/cm. for the auxiliary field which is necessary to produce any influence. If this minimum is genuine, one would expect it to appear also in connection with the "flashing" above mentioned, but it has not been observed.

It has been suggested by Schleede,[16] that the Lenard "phosphor" or center of luminescence is a foreign metal atom which has replaced a metal atom of the main mass of material in a crystal structure. The distinction between phosphorescing and fluorescing materials is attributed to the imperfection or distortion of the crystal lattice in the former case. This idea is supported by the fact that in general inorganic

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[12] Darwin, *P. N. A. Sc.*, Feb., 1923.

[13] Schmidt, *Ann. d. Phys.*, No. 3, 1923, p. 161.

[14] Gudden and Pohl, *Z. f. Phys.*, 1, p. 365, 1920.

[15] Herweg, *Z. f. Phys.*, 16, p. 29, 1923.

[16] Schleede, *Z. f. Phys.*, 1, p. 109, 1923.

solids which are made by melting and relatively sudden cooling are phosphorescent, whereas those formed by simply heating without melting or by crystallization without melting show only fluorescence. Schleede's own X-ray examinations of two different specimens of zinc sulphide indicate a marked departure from a uniform lattice in the case of the strongly phosphorescent form, while the fluorescent form was more regular. In this connection, one may recall that interesting class of fluorescent substances (ruby, certain fluorites, etc.) which particularly at low temperatures give extraordinarily sharp fluorescent lines. So far as I know, the only solids giving such sharp lines are certain crystals, and this suggests the same relation between structure and luminosity as has already been pointed out in the case of infra-red reflection, the sharpness of the lines being due to the extreme regularity of the surroundings of the emitting centers. A connection between structure and breadth of emission bands would seem to me as much to be expected as one between structure and duration of emission. We have recently been studying at Wisconsin the fluorescence of the ruby, and find another influence of structure—namely, a difference in the light emitted according as one examines the components (electric vector) vibrating along or perpendicular to the crystal axis, the regular structure either being responsible for the asymmetric constraints of the emitting center, or else permitting the asymmetric characteristics of the molecular center to appear.

The property of photoconductivity, or decrease of electrical resistance under the action of light, which is probably connected more closely than we at present understand with the fundamental optical and luminescent properties, appears to involve so many variables that it is almost impossible to decide what are the really significant factors. There seems little doubt that it is a case of conductivity by electrons, a larger number of which are made available, or set "free" or "loosened up," by the action of the light. It is observed with single crystals, aggregates of small crystals and amorphous material. In general, the photoconductivity as obtained with monochromatic light of various wave lengths but constant energy, shows a characteristic dispersion curve with one and sometimes two sharp maxima, though the form of this curve may vary with the intensity of the exciting light. Coblentz has recently[17] demonstrated that Ag2S has different photoconducting properties according as it is in the isometric (argentite) or orthorhombic (acanthite) form, but he has not connected this with any corresponding change in optical or other properties. Whatever connection there may be with the optical properties is apparently not simple[18] since for all substances so far examined in this way (diamond, ZnS and HgS), though the photoconductivity maxima come in the spectral region where absorption is beginning to increase rapidly, and the sensitivity falls nearly to zero with further progression into the absorption region, this sharp decrease occurs for wave lengths for which the materials are still quite transparent. Curves showing the photoconductivity current per unit of absorbed energy plotted against wave length, are practically linear for wave lengths longer than that giving maximum sensitivity, and this leads to the point of view that the absorption of light energy in solids is a quantum phenomenon, of a peculiar kind, however, since every quantum below a certain critical value must be supposed to liberate an electron or at least put it in condition to follow an external field. In the case of HgS and ZnS it even appears that the photo-currents are, within the experimental errors, quantitatively accounted for by this hypothesis. This is an interesting point of view, which, however, as yet rests on meager evidence obtained from measurements in that part of the spectrum where the photo-sensitivity and the absorptive power are both small and therefore relatively difficult to measure. It has been maintained that the liberated electrons came from the metal atom in the salt, but the evidence for this is hardly conclusive. There is, however, (Coblentz) in many cases a quite systematic shift of the sensitivity maximum with a change in the atomic weight of either the metal or the acid element in the salts. Gudden and Pohl[19] have also called attention to a systematic increase in the number of light sensitive salts which can be formed with the following series of anions

$$F^- < NO_3^- < SO_4^{--} < CO_3^{--} < Cl^- < Br^- < J^- < CN^- < O^{--} < S^{--} < Se^{--}$$

They find no photo-sensitive fluorides, nitrates or sulphates, and the number of sensitive salts increases as one passes to the right in the series, though cyanides form an exception to this rule in forming *no* sensitive salts. Some additional interest attaches to this because Fajans[20] from some general and rather vague considerations of the relation of structure to various properties, concluded that the "ease of distortion of the outer electron layers" increases regularly from left to right in the above series of anions.

The conductivity induced by light, in which the carriers are undoubtedly largely, if not entirely, electrons, is superposed upon a much smaller natural conductivity, which with equal certainty may be considered electrolytic in nature. There seem to be present in non-metallic crystals a number of ions

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[17] Coblentz, *B. S. Sci. Papers*, No. 446, 1922.

[18] Gudden and Pohl, *Phys. Z.*, 23, 1922, p. 417.

[19] Gudden and Pohl, *Z. f. Phys.*, 16, 1, p. 42, 1923.

[20] Fajans, *Die Naturwissenschaft*, 11, 10, p. 165, 1923.

which are able to follow an applied electric field, both the number and mobility of these ions being in general increased by an increase in temperature. According to Joffé[21] there are two fairly distinct types of crystals, one exemplified by calcite ($CaCO_3$), in which the conductivity is due to a relatively large number of positive ions of small mobility, and the other by quartz (SiO_2) in which there are a very small number of ions of much greater mobility, and of both signs. Foreign non-isomorphous ions present in a crystal are apparently largely "free," that is, in condition to follow a field and may be to a considerable extent removed by electrolysis, with consequent diminution of the conductivity toward a well-defined limit characteristic of the "pure" crystal. If the foreign ions are isomorphous, there is again a rapid diminution of conductivity produced by the passage of a current, such as is not found with "pure" crystals, but the limit approached seems to be between the values characteristic of the two constituents in the mixed crystal. Joffé concludes that, though the foreign ions are isomorphous, the lattice structure is less regular because of variations in concentration of the foreign ions from point to point and hence there will initially be more free or dissociated ions until, due to diffusion and current flow, a uniform condition is attained. It is possible not only to remove by electrolysis foreign ions which, as in the cases just mentioned, were embodied in a crystal during the process of formation, but also, as has been known for many years and recently studied in detail, to pass through a crystal ions from an electrode. The details of these processes are not clear, and there is room for much more study, which especially if combined with more detailed X-ray analysis, should throw light on the actual condition of what we have called the "free" ion, and the process by which it passes through a crystal—whether by a series of substitutions with the normal lattice ions, or through the interstices of the lattice. Joffé favors the latter idea and suggests that it may be possible to obtain estimates of ionic diameters by sifting them through crystals of varying mesh.

The processes we have just been discussing and the terminology we have been using suggest that this is the appropriate place to pass from the consideration of those substances primarily dielectric in nature to those primarily conducting and metallic. Metals are apparently always crystalline in structure, except for superficial and intercrystalline (Beilby) layers whose amorphous character is a matter of inference rather than direct proof, but the crystals are in general minute and variously oriented. There are, as always, two distinct problems—the problem of the single crystal and the problem of the crystal aggregate. Because of the difficulty of obtaining single crystals of suitable size, very little work has been done with them.

The most characteristic feature of metals is their high electrical and thermal conductivity, and largely through the extraordinarily skillful work of Bridgeman, there has been much recent progress in the study of these properties. Of all Bridgeman's work, covering as it does the pressure coefficient of electrical and thermal conductivity, the pressure coefficients of thermal e. m. f. and Thomson and Peltier heats, the departure from Ohm's law with high current densities, and the effect of tension on electrical conductivity, the most interesting in some respects is that dealing with the pressure coefficient of resistance. These results brought out the fact that of the thirty odd metals so far studied, there are six elements (Bi, Sb, Cs, Li, Sr, Ca) which have the surprising characteristic of increasing their resistance under pressure. One of these, caesium, has this abnormal behavior only in a high pressure modification, and as caesium is the most compressible of the metals, Bridgeman surmises that other metals might have abnormal forms under sufficiently high pressures. Particular interest attaches to all this from the standpoint of the formulation of a theory of the mechanism of electrical and thermal conductivity, to take the place of the original free-electron gas hypothesis, which we all use because of its simplicity, though it has been shown to be untenable by several lines of attack. The six elements showing abnormal pressure coefficient obviously require special treatment, but unfortunately these do not all behave alike as regards their tension coefficient, so that finally Bridgeman has been forced to include three more or less distinct pictures, the common basic idea being the conduction of both electricity and heat by the drift of a small number of electrons whose free paths are limited in different ways by atomic vibrations. In normal metals the electron is supposed to drift through the substance of the atoms themselves, which are arranged in chains of varying length, the free paths being determined largely by the amplitude of atomic motions; in the bismuth type, showing both abnormal (negative) pressure and tension coefficients a special hypothesis is necessary as to the law of force between atoms such that the amplitude of vibration will increase as the distance between atoms decreases; in the lithium type, having negative pressure but positive tension coefficients, the electrons are supposed to pass through the lattice between the atoms, and finally for strontium a combination of the two mechanisms seems necessary. It is necessary, also, to allow a varying amount of heat conduction by atomic impact, in addition to that resulting from the drift of electrons.

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[21] Joffé, *Ann. d. Phys.*, 72, No. 22, p. 461, 1923.

Another theory has been developed by Hall[22] which again avoids the specific heat difficulty by assuming a small number of free electrons and long free paths, but involves the rotations of both atoms and ions. They both have a thermal rotational energy proportional to the absolute temperature but of a much smaller order of magnitude than the translatory energy, and in addition the ions orient themselves in the external field. In this way, the drift of electrons from atom to ion (and so on) depends on the thermal motions and the field, and electrical conduction occurs in part by the motion of free electrons, but in larger part by the passing on of electrons from atom to ion. As E. B. Wilson[23] says, this theory has at least the advantage of giving the ions something to do, and as he further points out, Perkins's[24] observation that a negative charge diminished and an added positive charge increased the conductivity of graphite but produced no observable change with gold, are in qualitative agreement with Hall's hypothesis. This follows from the fact that adding electrons would probably reduce the number of ions and hence reduce the proportion of the conductivity due to ions—which is the preponderating part. How much importance attaches to this can not be decided until more extended observations on the effect of static charges are at hand.

The latest attack on this problem by Sir J. J. Thomson[25] puts the burden of both the electrical and thermal conductivity on the electrons, but the density of these is definitely fixed by the number and valency of the atoms. A certain type of crystal lattice is assumed for the metals, in which the "disposable" valence electrons are on lattices symmetrically placed with respect to the atomic (ionic) centers so that they are not bound to any particular atom, this being rendered plausible by computations of the compressibility of calcium. Consideration of the natural frequency of displaced single electrons as compared to linear groups or chains of electrons displaced along their length leads to the adoption of the latter as the unit of transportation. Having only one degree of freedom and containing many electrons (perhaps 10,000) equipartition applied to the chains as units does not lead to conflict with specific heat data. The chains move as rigid bodies in electrical conduction, but in thermal conduction there is also energy transmitted along the chain. A "collision" in this case is the passage of a linearly moving electron past the point of nearest approach to an ion, and there is no "persistence" of the velocity acquired by an electron in the applied field, except at very low temperatures when, since the time of passage of an electron by an ion may equal many periods of atomic vibration, there will be little transfer of energy to the ion, and superconductivity will appear.

Valency also plays an important rôle in the theory recently put forward by Waterman[26], in which the expression for the conductivity is the same as that derived from the simple electron-gas theory, the controlling factors being the mean free path and the concentration of the electrons, but the mean free path is assumed to be nearly constant and the variations in the conductivity are brought about by changes in the number of electrons per unit volume. This concentration of electrons is determined by applying the law of mass action to the reaction atom $\rightleftarrows$ ion $+ \nu$ electrons, where ν is the valency. The theory develops an interesting relation between valence and the temperature variation of conductivity which is quite consistent with the behavior of the alkali metals, and also a connection with the photo-electric and thermionic work functions for which there is some experimental justification though, as we shall see, the latter are at present rather uncertain quantities to tie to.

With respect to none of these theories have I attempted to mention all the characteristics of metals which in a more or less qualitative way are correlated by them, nor shall I attempt to criticize them or estimate their relative value. They are in the stage of working hypotheses, and having been developed from different viewpoints, each is most successful in its own field, and all will doubtless contribute to the ultimate clearing up of this very complicated problem.

One can anticipate most interesting developments from studies of single crystals, which will enable one to separate the properties of such crystals from those due to the state of aggregation. Recent measurements of the thermal conductivity of single bismuth crystals by Kaye and Roberts[27] show that the conductivity computed from the observed crystal constants, assuming a random distribution of crystals, agrees very closely (1 per cent.) with that observed for cast bars, provided no allowance is made for the effect of the contact between different crystals. It will be very interesting to find out whether the electrical conductivity of mass metals is connected in an equally simple way with the constants of single crystals.

We must now make our way out of these somewhat confusing internal conditions, where we have gradually become surrounded by lattices, amorphous layers, chains of atoms, chains of electrons, rotating dipols, free electrons which should not move and bound ones which will not stay still, to the region outside which we optimistically call a vacuum, and from this van-

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[22] Hall, *P. N. Acad.*, 7, No. 3, March, 1921; October, 1922.

[23] E. B. Wilson, *P. N. Acad.*, 9, p. 135, 1923.

[24] Perkins, *J. d. Phys.*, July, 1923.

[25] J. J. Thomson, *P. M.*, April and October, 1922.

[26] Waterman, *P. R.*, 22, Sept., p. 259, 1923.

[27] Kaye and Roberts, *P. R. S.*, 724, p. 98, 1923.

tage point consider some of the surface properties of solids. To begin with, we must give up the natural but entirely unwarranted expectation of finding conditions any simpler, since we have really added a few more variables to the already extensive list. One might naïvely think that the simplest surface phenomenon would be the passage of electricity, and in particular negative electricity, across the boundary of a metal, so let us begin with this, and in its simplest form, namely, the removal of electrons by the application of high electric fields. Early attempts to establish a definite minimum potential gradient, characteristic of a given metal by which electrons could be removed, were discordant, undoubtedly because of unidentified variations in the surfaces, more especially contamination by gases. Modern methods of outgassing have, however, been used in two recent instances. Using highly polished molybdenum spheres 1 cm. in diameter, with a minimum gap of 3 mm. Hayden[28] observed, after two days heating and exhausting, a maximum critical gradient of 1235 Kv/cm. More recently Millikan and Eyring[29], using heat-treated tungsten surfaces, observed a critical gradient of 800 Kv/cm. The order of magnitude is in satisfactory agreement—whether the difference is characteristic of the two materials or determined by slight surface peculiarities can not yet be said. One would anticipate that all spurious effects (except insensitive current measuring instruments) would tend to lower the observed critical gradient, and it is therefore somewhat surprising to find the highest values reported by Hoffman[30] for he worked in poor vacua (.001 mm.) and did not outgas his surfaces. His maximum value of 4800 Kv/cm was obtained with platinum-iridium surfaces, but lead gave 2200 Kv/cm. Hoffman worked with very small separations (1/3 to 3 wave lengths) and low voltages and thus avoided gas ionization troubles, but his high values are very likely due to the effect of gaseous double layers, and are not characteristic of the metals. Repetition of experiments on critical gradients and field currents, using crystal faces instead of artificially polished ones, might lead to more uniform results, and also, if faces having different atomic densities could be used, interesting results as to the "penetration" of external fields into the crystal lattice. One might expect the metals to arrange themselves in the order of their thermionic work functions, as regards the difficulty of removing electrons, but the results for molybdenum and tungsten are in the reverse order. One might also expect that a considerable increase in temperature might lead to a lowering of the critical potential gradient because the thermal motions of the atoms would on the average "open up" the surface and leave "holes" for the easier escape of electrons. Millikan reports that an increase in temperature from 300° K to 1000° K produces no change in the field currents (and presumably none in the critical field) but that at 1100° K the field currents are slightly larger, which he attributes to increased penetration of the field, possibly due to the development of minute cracks.

One may substitute for these gross external fields the molecular fields of another proximate surface and here again we find great differences and discrepancies in the results, which just now seem by way of being somewhat cleared up[31]. Evidence is accumulating for the truth of Coehn's law that the amount and sign of the charge assumed by a substance A when touched or rubbed by another, B, is proportional to the difference of their dielectric constants. Steel fits into the series of dielectrics if its dielectric constant is taken as 3.1. While, as Richards says, this may be a reasonable value for the constant expressing the behavior of the bound electrons in a metal, it is not clear why the dielectric constant should be the controlling factor in the present case, whereas in all other cases (except the reaction toward infra-red waves less than 4 μ long) the bound electron effects in metals seem to be masked by the behavior of the "free" electrons. The charges produced by the impact of a dielectric sphere on a metal are considered by Richards as due to the true volta contact effect just mentioned and an effect of electron inertia such as was discovered by Tolman and Stewart. One might also expect a difference between impact and mere contact or rubbing because of the breaking down or violent distortion of the structure of both surfaces occurring in the former case.

The old conflict as to the essential nature of the volta contact e.m.f. between metals has subsided, with the idea that it is an "intrinsic" property triumphant over its "chemical" rival. The "intrinsic" hypothesis is of course nothing more than a name, since we have as yet only the vaguest ideas as to the cause of the hypothetical natural attraction of metals for electrons, and its present favor is due in large part to the intimate connection which has developed between contact e.m.f. and thermionic and photoelectric discharge. The primary difficulty in all measurements of this sort is the purification of the surface—particularly from gas contamination. Without question no one has ever worked with gas-free surfaces, and probably no one ever will, but it is becoming more and more possible to obtain *reproducible* surfaces, and whether or not they contain some gas is after all

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[28] Hayden, *J. Am. Inst. Elec. Eng.*, No. 11, p. 852, 1922.

[29] Millikan and Eyring, *Phys. Soc.*, Sept., 1923.

[30] Hoffman, *Z. f. Phys.*, 4, p. 363, 1921; 5, p. 109, 1923.

[31] Coehn, *Z. f. Phys.*, 5, p. 242, 1921; Richards, *P. R.*, Aug., 1923, p. 122.

of somewhat secondary importance provided we know what the actual condition is.

The latest most accurate work of Dushman[32] and Davisson and Germer[33] only serves to confirm the validity of both the first ($i = A_1 T^{\frac{1}{2}} e^{-\frac{b}{T}}$) and second ($i = A_2 T^2 e^{-\frac{d}{T}}$) Richardson thermionic equation, it being impossible as yet to discriminate in favor of either one of them. It is also impossible to decide at present as to the value of the constant A, though the indications are that it is a universal constant, as Dushman's theory would make it. One of the attractions of Richardson's theory is that it accounts in a very simple way for the energy of the thermions, since it is based on the idea of a free-electron gas in the metal for which the mean kinetic energy is the equipartition value $\frac{3}{2}$k T. This is, in fact, the most successful application of the electron gas hypothesis, where it has persisted in spite of difficulties encountered in other directions, but there is now a little thermionic evidence against the usually assumed value for the mean energy inside the metal. This is furnished by the work of Davisson and Germer, since it is found that the values of the work function for tungsten obtained by the compensated cooling method, and from the temperature variation of the thermionic current, are brought into excellent agreement not only with each other but with Dushman's and Lester's, if they are all handled on the assumption of zero kinetic energy for the electrons within the metal, but are not in good accord if handled on the usual assumption.

The hypothesis of zero internal kinetic energy removes the specific heat difficulty, but necessitates special assumptions to account for the initial energy of the thermions. Evidence of the insufficiency of the simple electron gas hypothesis may be furnished by observations such as those of Göetz,[34] wherein the thermionic discharge from electrolytic copper and iron and manganese was followed through various transformation points and the melting point. In terms of the first Richardson equation, Göetz records changes in either A or b or both constants, at the melting points and at three other temperatures, two of which correspond to known transformation points of iron. These interesting results can hardly be accepted as definite without further confirmation, in view of the great liability to gas contamination even with the prolonged outgassing which Göetz used. One would expect a different surface-gas equilibrium above and below the melting point, and a crystal transformation might also lead to a similar change. Richardson's observations[35] on the effect of hydrogen on the thermionic discharge from platinum and tungsten may, on the other hand, be taken as a count in favor of the electron-gas theory, since he was able on this basis to compute changes in the contact e.m.f. which were in fair accord with those observed, but the conditions of the experiments were of necessity so complex that the interpretation is, as Richardson himself says, somewhat uncertain. One may point out in particular that the application to this case of a generalization deduced from photo-electric measurements as to the constancy of the "stopping potentials" would lead to the conclusion that the changes observed were not in contact e.m.f. but in initial velocity of electrons. The present status seems to be, then, that there is no doubt that thermionic discharge is an intrinsic property of metals, which can be expressed with all the necessary accuracy by a simple exponential equation, possibly with only one constant characteristic of each metal, that it probably is not simply connected with the process of electrical conduction, and that the origin of the thermionic energy is uncertain. One's discouragement with this situation is perhaps somewhat relieved by the realization that a puzzling but apparently unescapable equation *can* be derived *without* the use of the quantum theory[36].

The situation with regard to the photo-electric effect is just now equally confused. It has been shown many times, most recently by Tucker,[37] that the photo-sensitivity of metals can be reduced almost, if not quite, to zero by prolonged heating or otherwise outgassing, but in every case the observations were open to the interpretation that there had occurred a shift of the long wave limit toward the ultra-violet beyond the spectral range of the apparatus, and not a vanishing of the photo-electric

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[32] Dushman, *P. R.*, June, 1923, p. 623.

[33] Davisson and Germer, *P. R.*, 20, p. 300, 1922.

[34] Göetz, *Phys. Z.*, 23, 18, p. 377, 1923.

[35] Richardson, *P. M.*, 43, p. 162, 1923.

[36] Heated metals are puzzling not only for their thermionic property—they are equally baffling as regards their radiation. Their spectral emission curves are similar to those of a perfect radiator, but none of the well-known laws will suffice even with changes of constants. Here again one is painfully conscious of the dangers of surface contamination. We have at Wisconsin an accumulation of data covering a wide range of temperature for a dozen different metals, taken under conditions as good or better than have ever been used elsewhere, but I have been unable to correlate the radiation properties to any other properties of the metals. For low ranges of temperature, the simple Drude relation between total emissivity and electrical conductivity is approximately verified—but this is only a first approximation and neglects factors which become increasingly important at high temperatures.

[37] Tucker, *P. R.*, Dec., 1923.

effect. We now have at Wisconsin an outfit in operation with which we expect to be able to follow the shift for platinum down to about 1500 Å, and Dr. Swann informs me that Mr. Woodruff at the Ryerson Laboratory is extending Tucker's work in a similar way and has shown that Tucker's vanishing sensitivity *was* due to a shift of the long wave limit. How far this limit can be shifted by the most intense and prolonged outgassing remains to be seen. Again, Suhrman[38] after long continued heating finds a long wave limit of 2700 Å for platinum, but he is of the opinion that the real limit for the gas-free metal must be much farther in the ultra-violet, because of the effect of outgassing on the sensitivity to wave lengths from 2400 to 2150, much shorter than the apparent limit[39]. It is evident that there is still great uncertainty as to the real photo-electric characteristics of platinum. In the meantime, Kazda, working with Millikan, has taken extreme precautions to secure a clean surface of mercury, which was continually flowing and being replenished by distillation, and for such a surface he finds a definite long wave limit at 2735 Å. This ought to be as nearly a gas-free surface as has ever been worked with. There is also good reason to consider the long wave limit (2700 Å) obtained in the same laboratory by Neilsson for aluminum melted in vacuum as a limiting constant for this metal. If one could accept Suhrman's result of 2700 Å for platinum, it would begin to look as if all gas-free metals had the same long wave limit—a most improbable result in the light of thermionic values for the work function. But Suhrman's wave length is undoubtedly too long, and it is probable that in the end, definite and characteristic long wave limits will be found for each metal.

The complexity of the situation was great enough before Shenstone[40] confirmed his earlier observations of a change in the photo-electric sensitivity due to the passage of an electric current through the metal under examination, as observed in a very high vacuum. The current has a direct and rapid effect when first thrown on, which more or less slowly (in from one to many hours) disappears after the current is cut off, the effect of a given current being the same whether it is applied suddenly or gradually attained by successive increases. In general (for Bi, Cu, Ag, Ni, Au, and Pt) the effect of the current is to increase the photo-sensitivity, though with copper and gold a maximum is quickly reached and the sensitivity falls below its initial value, the cycle of changes produced by increasing and stopping the currents being quite accurately reproducible. Shenstone has measured the small temperature changes involved (of the order of 5° to 30° C.), and while the reduction of these temperature changes by a cooling device modified the characteristics of the effect, it did not tend to eliminate it. It is natural to attribute these very interesting results to changes in surface gas content, which Shenstone has of course carefully considered, and it must be admitted that this explanation is hard to reconcile with the cycle of changes through which copper could be repeatedly carried. For the present, we must leave it as a probable indication of some change in surface structure produced by the passage of an electric current parallel to the surface, rather than as evidence of an orientation phenomenon directly associated with electrical conduction, for the latter would hardly be expected to show such prolonged after effects.

The Shenstone effect is undoubtedly involved in the changes in the photo-electric sensitivity of platinum observed by Wello,[41] which were produced by successive short heatings of platinum foil to gradually increasing temperatures, but outgassing effects were also certainly present and probably predominant at the higher temperatures. There is much to favor Hallwachs's view that we should consider a loosely attached gas layer as well as the gas content of the metal itself, or some similar hypothesis of a surface content and volume content of gas which are affected differently by increase in temperature. The surface gas condition will be controlled by the volume density of gas in the metal, the rate of diffusion of this gas to the surface, the rate of escape of gas from the surface and the amount of gas in the surrounding space. All the observations on photo-electric effect and contact e.m.f., except possibly Shenstone's, seem to fall in with such a point of view.

There is one point on which observers seem to agree, perhaps because no one has directly investigated it for several years, namely, that the retarding potential necessary to stop the photo-electric discharge, with a given wave length of light, is the same for all metals examined. From this Millikan concludes[42] that either the energy of the photo-electrons must be hν *after* their escape from the atom, or else that the photo-electrons are initially the "free" electrons rather than the atomic or bound ones. The only reasonable way of accounting for the first alternative is to say that the energy absorbed by an electron in an atom is just enough to get it out of the atom and leave it with a kinetic energy hν which is the conclusion reached by Barkla in discussing the photo-electric effect of X-rays, and which associates the whole phenomenon intimately with atomic structure. Spectroscopy has familiarized us with many

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[38] Suhrman, *Ann. d. Phys.*, 372, p. 43, 1922.

[39] Suhrman's curves suggest that he may have been bothered with scattered light of short wave length.

[40] Shenstone, *P. M.*, May, 1923, p. 918.

[41] Welo, *P. M.*, March, 1923, p. 593.

[42] Millikan, *P. R.*, Sept., 1921.

quantum changes of atomic energy, but there is nowhere any analogy to this case, where the *surplus* energy equals hν. The second hypothesis according to which the photo-electrons come from the mass structure rather than the atomic structure is simpler, but it throws the responsibility for the kinetic energy quantum upon the absorption by a free electron, and for this there has never been any hypothesis except the original Einstein one of point quanta.

Whittaker[43] not long since suggested a very interesting atomic model, especially designed to account for the interchange of energy by quanta ($\Delta E = h\nu$) between an atom and an electron. The essential feature is a magnetic structure which is started in rotation, thus producing a magnetic current, by the absorption of radiation. If the absorbed energy just amounts to hν it follows from the equation of the model that an approaching[44] electron will be sucked through the atom and sent off with the kinetic energy hν. The model also takes care of the conversion of the energy of inelastic electron impact into radiation in accordance with the quantum law, and can be made to involve the general spectroscopic characteristics of the Bohr atom, but it does not involve any mechanism for the absorption of radiant energy. The Whittaker mechanism seems somewhat clumsy, but it may of course be taken as merely illustrative, and on the other hand we may recall that rival models escape this criticism by suggesting no mechanism whatever.

I have already referred to the work of Cario and Franck as affording experimental evidence for the occurrence of a particular type of impact in which the energy of an excited atomic state is transferred to another atom. If we assume that this absorbed energy ($\Delta E = h\nu$) may also be transferred by impact to a "free"[45] electron, the photo-electric requirements would be satisfied. It would be an inner and not a valence electron which would be responsible for the absorption, since the latter are probably much altered in the solid state, forming the "free" or at least loosely attached structural electrons. There is difficulty in picturing the absorption of all frequencies—but this may be thought of as a statistical effect due to the varying distortion of the outer levels of the atom in the course of the constrained thermal motions.

Whatever detailed hypothesis we may adopt, the constancy of the stopping potentials leads to the conclusion that the energy expended by the electron in passing through the surface is $h\nu_0$ where ν_0 is the low frequency limit, and one would expect an agreement between the surface work functions determined photo-electrically and from thermionic observations, properly corrected for temperature change. All that can be said is that these two quantities do agree as to order of magnitude, and until the uncertainties surrounding the photo-electric long wave limit can be cleared up, this is as much as could be expected.

Since we are entirely uncertain regarding the fundamental point as to how the light acts upon the photo-electron, speculations regarding this are of interest. One such is by Bothe[46], based on the point-quantum idea, who points out that in the impact of a quantum with an electron, if there is to be conservation of energy and linear momentum, then the quantum can not *disappear* (*i.e.*, transfer all its energy to the electron) if the electron is free. From this he concludes that the photo-electrons are initially bound, and discusses the impact and annihilation of a quantum under these conditions, especially as regards the direction of motion of the liberated photo-electrons. He computes the ratio of the forward- to the backward-going electrons produced by different wave lengths in gold, and these prove to be in the proper sequence but all larger than the observed ratios, while for γ-rays the computed asymmetry ratio is much smaller than that observed. This last is a discrepancy which can not be attributed to the scattering of the photo-electrons in the metal, since this would always tend to diminish the asymmetry; and the situation is only partly relieved by attributing rotational energy to the light quantum.

From what has been said it seems evident that a summary statement of the status of the photo-electric problem must be at least as vague as that applied to the thermionic effect, and indeed it may be put in almost the same words: the experimental verification of a simple but baffling equation ($\frac{1}{2}mv^2 = h(\nu - \nu_0)$), great uncertainty as to the values of the constants involved, no known connection with electrical conductivity or with optical properties, and only vague ideas as to the source of the energy of the emitted electrons.

As a final method of removing electricity from metals we may consider the bombardment of a metal surface by electrons. If these are of relatively high

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[43] Whittaker, *P. R. S. E.*, 52, 2, p. 129, 1922.

[44] The question of the probability that an electron would approach just as the energy reached the value hν would seem to require some consideration.

[45] It is not unreasonable to consider the "free" electrons in metals as of fundamental importance in the photo-electric effect, in spite of the known photo-electric discharge from insulators, for we have seen that light evidently loosens up the structure of insulators, putting electrons in a more or less free condition. The photo-electric effect in insulators may therefore involve two somewhat distinct processes.

[46] Bothe, *Z. f. Phys.*, 7, 2, p. 137, 1923.

velocity they would be expected to penetrate into the actual atomic structure, and indeed, in the skillful hands of Davisson and Kunzman they have presented us with the most striking and direct evidence we have for the nuclear theory of the structure of the atom. Low velocity electrons will undoubtedly tell us something about the mass structure, though as yet the conclusions are indefinite. Since the outer atomic levels are, without doubt, seriously altered in the mass (otherwise atomic optical properties would show themselves) it is improbable that the atomic ionization and resonance potentials will be observed in connection with low velocity bombardment—though there might well be, in some cases, observable effects due to analogous quantum changes, which might be expected to be more pronounced in those metals where the outer electron levels are the least disturbed by aggregation.

Mention of the subjects of this address would, I suspect, bring to most of us first a thought of the wide variety and complexity of the experimental and theoretical methods involved and then a feeling of the apparent disconnection between them, as if each group of workers were so intensely interested in cultivating their own field that they paid but scant attention to what was being done in others. Even an incomplete account such as the present may be of value in showing that while the complexity is real the impression of disconnection is largely superficial, and in pointing out some of the cross-relations which are developing between the various methods of attacking the fundamental problem of the constitution of solids. If in this presentation I have appeared to overemphasize the failures of our theories and the defects of our experimental knowledge, you must accept this as in part a matter of temperament. The pessimist dwells on successes to keep his courage up —the optimist enjoys facing things as they are. On the other hand, it is the failures and the inconsistencies of our interpretations which point the way to new knowledge, and it is in one very real sense true that a theory is valuable inversely in proportion to its apparent completeness and rationality. I have no regrets, therefore, in commending to you the subjects of this afternoon's discussion as ones in which there is great room for improvement.

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THE ORGANIZATION OF SCIENTIFIC MEN

In discussing altruism and the struggle for existence[1] David Starr Jordan has urged that those who band together win. Those who can stand shoulder to shoulder loyally have the promise of the future. Those who can not hold together find every man's hand raised against them.

Some contemporary developments are eloquent that the traditional policy of individualism which animates so many intellectual workers is harmful to the best interest of the workers, their work and of society, and that it must ultimately be abandoned and replaced by a policy of cooperation. There is scarcely justification for an absorption in one's specialty which leaves no time for consideration of the welfare of one's family and fellow workers.

We believe that certain organizations already in the field are doing valuable work in this direction, though most of them are handicapped by limitations greatly reducing their effectiveness. To mention a few only: The social and scientific activities of the American Association for the Advancement of Science are considerable, but its contribution on the economic side is very small; the scientific-technical branch of the Washington, D. C., local of the National Federation of Federal Employees did a valuable piece of work in behalf of the reclassification of the civil employees of the government, but has now become inactive, and is, of course, limited to employees of the United States government; in their own spheres of activity such organizations as the American Medical Association and the American Association of Engineers have accomplished much, but their contributions have been very largely to the welfare of the more or less homogeneous groups which they represent; more important, perhaps, than any of these (at least for university men) is the American Association of University Professors, which, however, is limited to certain grades of university men, and which has not been nearly so effective as it might be with a larger membership and more adequate dues.

It seems clear that a professional organization more inclusive than any of these is needed, embracing workers in *universities, academies, museums, private scientific* and *intellectual foundations,* and in the *service of state* and *federal governments,* to insure the social and economic welfare of the intellectual worker and to increase the value of his contribution to society. Business interests and labor have long had their organizations, through at least some of which they secure effective cooperation. Even the long-suffering farmer is taking some elementary lessons in the fine art of mutual endeavor. Why should the intellectual worker lag so far behind? In some of the countries of the old world, perhaps as a result of an almost insupportable economic pressure, there are signs of an awakening. A writer in a recent number of the British Medical Journal[2] reports that there are, in

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[1] "Footnotes to Evolution," 1913, p. 28.

[2] Quoted in Science, May 18, 1923.