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1 June 1953

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ELECTRONIC STRUCTURE OF METALS AND ALLOYS

At the spring meeting of the Physical Society, held at Leeds University on March 30 - 31, some recent successes of the collective electron treatment-band theory, as applied to metal and alloy systems, were discussed. Dr. F.E. Hoare (Leeds) first described his magnetic and specific heat studies on the Pd-Ag alloys as a function both of temperature and of Ag concentration. Measurements of these quantities have been made over the range from liquid hydrogen temperatures to room temperature, the magnetic susceptibilities being measured with a ring balance of the Sucksmith type. The shape of the band,  $N(E)$  vs.  $E$ , determined from measurements of the specific heat on a number of alloys with differing percentages of silver, was found to be concave upward, as is shown in Fig. 1(a). This shape was sufficient to predict the observed maximum in the curve of magnetic susceptibility vs. temperature, Fig. 1(b). Hoare has been making similar studies of the Pd-Rd and Pt-Ir systems.

Dr. P. Rhodes (Imperial College, London) discussed the more general theoretical problem of finding the relationship between magnetic susceptibility and band shape. He assumed a band shape of the form  $N(\xi) = a + b\xi^n$ , where  $\xi$  is measured from the top of the band as shown in Fig. 1(a). For the case of  $n > 1$ , an important parameter was found to be  $\phi = (b/a)\xi_0^n$ , where  $\xi_0$  is the width of the unoccupied part of the band. The case  $\phi = 1.146$  gave the most pronounced maximum in the curve of magnetic susceptibility,  $\chi$ , versus temperature, whereas no maximum was found in the curve where  $\phi \geq 5$ .

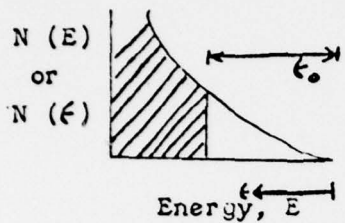


Fig. 1(a) Density of States Curve

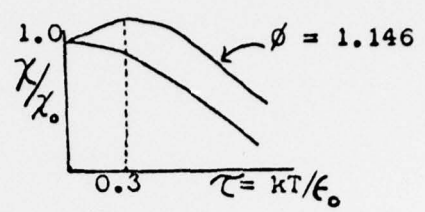


Fig. 1(b) Magnetic Susceptibility versus Temperature

Dr. E.W. Elcock (Leeds) discussed the magnetic properties of polyvalent metals, such as Zn, Cd, Bi, and Bi-Pb alloys, as calculated from a simple band model. He considered the overlapping set of bands shown in Fig. 2(a) which are simplified for the calculation by cutting them off along the dotted lines; since the principal contribution to the magnetic susceptibility comes at the Fermi surface, the cutting-off procedure should not introduce appreciable error. Elcock obtained a number of different  $\chi/\chi_0$  curves, such as are shown in Fig. 2(b), depending on the position of the Fermi surface. His results show that where two bands overlap, and where the almost full lower band has a steep parabolic head, then almost any type of  $\chi$  versus  $T$  behavior is possible; thus the very different  $\chi - T$  curves which are observed for the metals mentioned above can be explained.

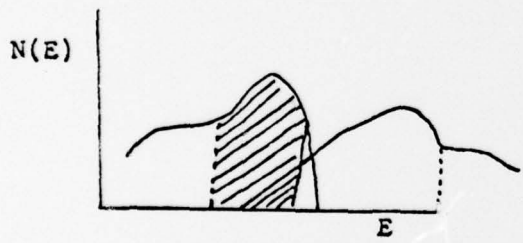


Fig. 2(a) Overlapping Band Scheme for Polyvalent Metal

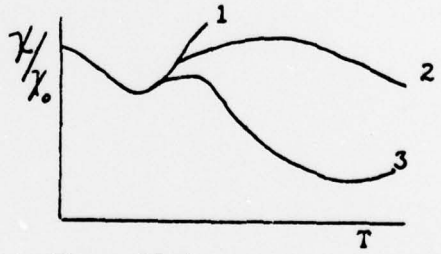


Fig. 2(b) Magnetic Susceptibilities as Calculated from Bands in 2(a)

Elcock has been particularly interested in the Bi-Pb alloys. He has obtained a  $\chi$ -T curve with a single broad maximum characteristic of Bi + 1.04%oPb; he next varied the Fermi surface, i.e. the number of electrons per atom, by fixed amounts from that characteristic of Bi + 1.04%oPb, and obtained  $\chi$ -T curves which agreed very well with the measured data for these alloys.

#### TRANSPARENT ELECTRICAL CONDUCTING COATINGS FOR GLASS

Dr. G.A. Veszi of Megatron Ltd., London, exhibited a series of transparent electrical conductive coatings for glass at the Physical Society Exhibition held in London on 13 - 17 April. One group of these coatings has been developed as a result of his observation that the electrical conductivity of gold can be greatly enhanced if it is evaporated on to a substrate of bismuth, antimony, or lead oxide. An additional layer of the oxide is put on top of the gold. He has found Bi<sub>2</sub>O<sub>3</sub> to yield the best results and can get a surface resistivity as low as 5 ohms per square with a visible transparency of 75 per cent. The oxide films are sputtered from electrodes of the pure metal in an atmosphere of argon containing a small amount of oxygen.

An even higher transmission has been found in very recent experiments with indium oxide, but the surface resistivity runs in the vicinity of 1000 ohms per square. The indium oxide is sputtered in a single layer without any gold, and then heated at about 200°C.

#### AN INFRARED ELECTRO-OPTICAL SHUTTER

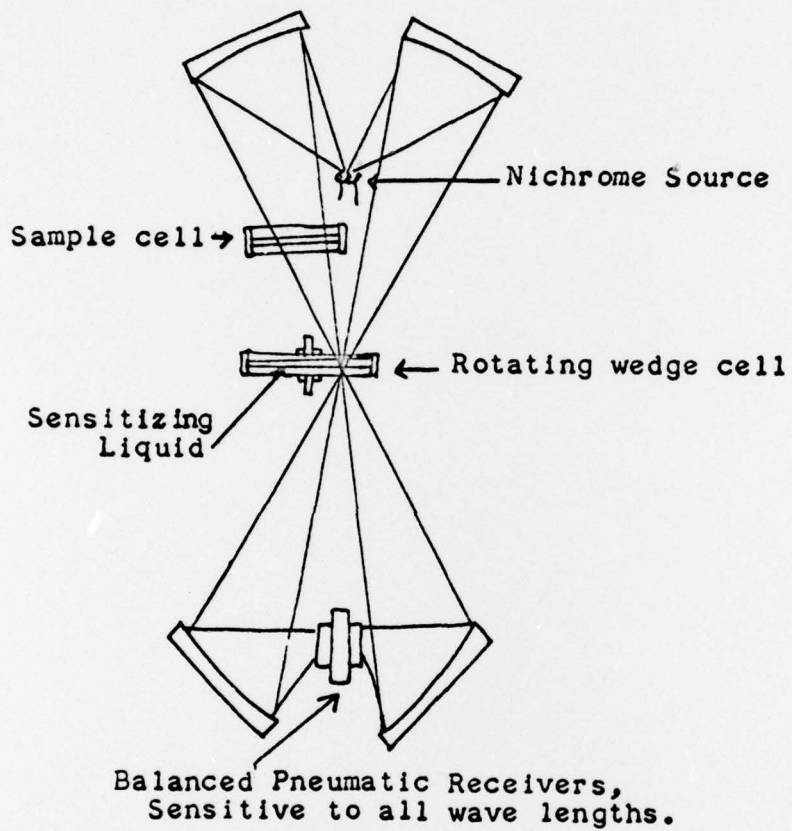
At the recent Physical Society Exhibition in London, Dr. A.F. Gibson of the Telecommunications Research Establishment, Great Malvern, Worcs., showed an interesting demonstration of the alteration of the optical properties of germanium carrying an electrical current. A block of germanium about 1 x 5 mm in cross section and 10 mm long was illuminated on one end (i.e., down the length of the specimen) with a focused spot of light from an incandescent source. Electrodes on the top and bottom surfaces served to introduce electrical carriers whose presence increased the infrared absorption of the germanium. The electrodes had a potential difference of 3 - 4 volts giving a current of the order of 20 milliamps.

In this demonstration the electrodes carried a signal derived from a phonograph, and the current served to modulate the light beam at audio frequencies. The transmitted beam was collimated by a mirror and aimed at a receiver 10 ft away, the receiver using a photoconductive cell of lead sulfide. The transmitted beam with its modulation envelope was shown to consist primarily of the near infrared wave lengths for which the receiver was most sensitive.

#### AN INFRARED LIQUID ANALYZER

Dr. K.F. Luft, Office Nationale d'Etudes et Recherches Aeronautiques, Paris, is constructing an infrared liquid analyzer whose design derives in some ways from the Pfund type positive filtering gas analyzer. One light source is used, a nichrome coil. The light from this is divided into two beams, one of which passes through the sample whose composition is to be determined; both of the beams then pass through a control specimen of the liquid whose presence is to be detected in the sample. This liquid is contained in a cell whose thickness varies with time to produce modulation of the wave lengths which it absorbs. The modulation of thickness of the cell is done by rotating a circular cell at about 10 cycles a second, the outer surfaces of the cell being plane parallel while the space in the interior, which contains the liquid, is wedge shaped (see figure). The two beams are then brought to a focus on two black pneumatic receivers connected in opposition. The instrument is balanced in the absence of the sample, and the resulting imbalance when the sample is introduced is a measure of the concentration of the unknown. The presence of any other liquid yields no AC signal because the wave lengths for which it is an absorber are not modulated. This instrument is now in construction and has not yet been tested, but it should prove valuable.

The receivers are identical with those used in the conventional gas analyzer except that they have been blackened to render them equally sensitive at all wave lengths.



INFRARED LIQUID ANALYZER

## FARADAY SOCIETY DISCUSSION ON SOLUTIONS OF NON-ELECTROLYTES

The Faraday Society Discussion on "Solutions of Non-Electrolytes" (London, 16 - 18 April, 1953) provided an opportunity for an exchange of views by most of the active workers in this field. As usual, it is planned to publish the complete proceedings in about six months' time.

Some of the new results presented are discussed briefly in the following paragraphs. A somewhat more detailed account of the discussion is given in Technical Report ONRL-49-53, available from the Technical Publications Office, Office of Naval Research, Code 740, Washington 25, D.C.

### Theory

Professor E.A. Guggenheim (Reading) has revised his opinions on the calculation of thermodynamic functions of solutions, and now feels that the zeroth order approximation in lattice theories is better than he believed in the past. He emphasized that not only the configurational terms but also the interaction terms may generally contribute to the entropy changes observed since they may be temperature dependent. The new system of integral equations for the distribution function presented by Professor J.G. Kirkwood (Yale) may lead to a consistent theory of phase transitions.

In an attempt to test the usefulness of the several modifications of the lattice theory of solutions, Dr. J.S. Rowlinson (Manchester) compared the corresponding theory of pure liquids with experimental results and concluded that only the most complicated model, in which all neighbors are considered, can be expected to form an adequate basis for a theory of solutions.

### Orientation Effects in Liquid Mixtures

Dr. J.H. Pople (Cambridge) presented a semi-quantitative treatment of the effect of orientation on the thermodynamic properties of liquids. The effect is significant and arises from additional cohesive energy and from a loss of entropy associated with hindrance of free rotation. The absence of free rotation in liquid carbon tetrachloride, and in liquid benzene, is indicated

by their high configurational heat capacities. This approach can be directly extended to liquid mixtures, and its usefulness was shown by comparison with the available data on the well-known benzene-cyclohexane, benzene-carbon tetrachloride, and cyclohexane-carbon tetrachloride systems. Recent experimental measurements on the heat capacities of a number of liquids and binary liquid mixtures were presented by L.A.K. Staveley (Oxford). The results obtained on mixtures of benzene with ethylene dichloride, carbon tetrachloride and acetic acid are all interpreted on the assumption that the benzene molecules rotate more freely in these mixtures than they do in pure benzene.

### Experimental

In introducing the experimental discussion Professor D.H. Everett (Dundee) emphasized the need for comprehensive experimental data on fairly simple systems covering the largest number of thermodynamic properties and temperature range. The usefulness of an agreement between experimentalists and theoreticians on the best methods of presentation of results to facilitate comparison between them, was briefly discussed.

In a contribution with J.L. Copp, Professor Everett dealt with amine solutions exhibiting both upper and lower consolute phenomena. The reasons for the relative rarity of lower consolute points in comparison with upper consolute behavior were discussed; the presence of lower consolute behavior in aqueous media is closely related to association between the components and to effects arising from the interaction of the inert hydrocarbon group of the second component with the water structure.

### Hydrogen Bonded Systems

An attractive, consistent interpretation of the behavior of water-hydrogen peroxide and water-alcohol mixtures was presented by Professor W.F.K. Wynne-Jones, (Newcastle). The thermodynamic properties are interpreted by assuming that the ordered structure of the water is reinforced by hydrogen peroxide but is weakened (and eventually destroyed) by alcohol. This is due to the fact that the hydrogen peroxide molecules can fit into the water structure but the alcohol molecules cannot do so because of their alkyl groups, and thus remain outside.

## Solubility in Compressed Gases

The discussion of solubility in compressed gases indicates a revival of interest in this somewhat obscure field of considerable industrial significance.

A.H. Ewald, W.B. Jepson, and J.S. Rowlinson (Manchester) gave a theoretical analysis in which the excess solubility is expressed in terms of a virial expansion; the successive coefficients represent the clustering of 1,2, etc. gas molecules around a given solute molecule. This theory provides a good qualitative explanation for the available data, including recent measurements in Manchester on solutions of iodoform in propane and in dimethyl ether. Their results indicate that the hydrogen atom of iodoform is capable of forming a hydrogen bond with the oxygen of the ether.

Dr. T.J. Webster (British Oxygen Co., Ltd., London) showed that it is difficult to interpret the available experimental observations, without assuming the formation of a solution in the gas phase of molecules which would otherwise condense. Referring to some of his own recent experimental results he cited some striking figures indicating how large these effects are: the concentration of carbon dioxide in air at  $-150^{\circ}\text{C}$  and 280 atm. is about 1500 times greater than would be calculated using the ideal expressions. An opposing approach to these systems was suggested by Dr. R. Strickland-Constable (Imperial College, London). He examined the observed solubilities in terms of the density of the phase in question and finds that these solubilities are quite low when the compressed gas is considered as an ideal liquid. Plots of the solubility of carbon dioxide in compressed air and in liquid air appear to be continuous, indicating that this approach may be of value. In an attempt to unify these views Professor H.C. Longuet-Higgins (King's College, London) suggested that the key concept is that of the cohesive energy which is a sensitive function of the volume.

## THE EFFECT OF ULTRASONIC RADIATION ON THE OXIDATION OF NITROGEN IN AQUEOUS SOLUTION

A systematic study was made in the laboratories of Professor H. Gobrecht (Technical University, Berlin) on the reaction between nitrogen and oxygen dissolved in water. It was found that above a minimum intensity, acoustic waves have a strong catalytic effect on the reaction. This was interpreted to support the view that

the reaction is intimately connected with cavitation in the fluid medium.

The reaction between nitrogen and oxygen in water was studied at 30°C using acoustic radiation of 350 Kc frequency. The course of the reaction was followed by measuring the increase in the electrical conductivity of the water as the reaction proceeds. The water is exposed to clean air while the irradiation occurs and it was found that there is a continuing increase in electrical conductivity suggesting that the reactants are rapidly and smoothly replaced in the liquid as reaction proceeds. It was estimated that the ammonia formed from the reaction between nitrogen and hydrogen (the hydrogen being formed by decomposition of the water) would have a negligible effect on the conductivity, compared to that of the NO and NO<sub>2</sub> formed.

The energy input is estimated from separate experiments in which the heat produced by the absorption of the ultrasonic waves in pure water, free of nitrogen, is determined. Experiments in which pure, nitrogen free water is irradiated under an atmosphere of oxygen and hydrogen showed no measurable increase in electrical conductivity.

In investigating the influence of the energy of the ultrasonic radiation on the reaction between nitrogen and oxygen, it is found that a certain minimum energy is necessary for any reaction to take place; above this energy the change in conductivity in unit time is a linear function of the energy. These observations are interpreted to support the view that such chemical reactions are intimately connected with cavitation in the liquid (cf. R.O. Prudhomme, Proc. Int. Conf. on Ultrasonics, Brussels, June 1951, p. 182). The effect of temperature on the rate of chemical reaction at different intensities of irradiation was found to give a family of straight lines converging to zero reaction at about 70°C. This can be logically explained under the assumption that above this temperature the solubility of the reactants in water is too low to permit any reaction.

#### EXPLANATION OF THE PORTEVIN-LE CHATELIER EFFECT IN ALUMI-NUM ALLOYS

Professor A.H. Cottrell (Department of Theoretical Metallurgy, University of Birmingham) has recently proposed a theoretical explanation of the Portevin-Le Chatelier effect. This effect has been observed at room temperature as serrations in the stress-strain curve of Al containing a few per cent substitutional impurities (see Fig. 1), the

serrations starting only after a certain amount of strain (0.1 to 1 per cent). The effect has been observed many times in aluminum which contains a few per cent of Cu, Mg, or impurities of the transition metals; it is not observed in pure Al. It appears most prominently in freshly quenched alloys when the alloying elements are retained in solution, and it has only been observed in a range of temperature in the vicinity of room temperature.

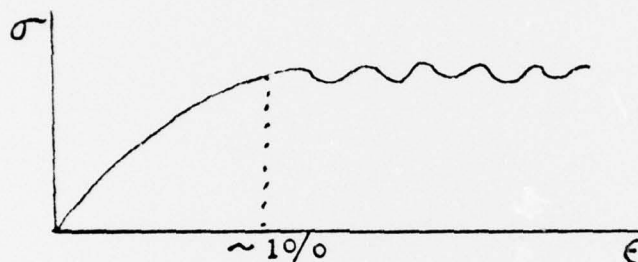


Figure 1

Serrations have often been observed for stress-strain curves on soft iron at temperatures around 200°C. In this case it is known that the serrations are due to repeated yielding which occurs because nitrogen and carbon atoms can diffuse to the dislocations fast enough to produce strain aging during the plastic deformation. It is attractive to assume that the Portevin-Le Chatelier effect has a similar origin with Cu or Mg as the active solute atoms for this case. The main difficulty in this explanation is to explain how the solute atoms can diffuse rapidly enough; their rates of diffusion in Al at room temperature are much too small. Cottrell's explanation is that the rate of diffusion is increased sufficiently by the concentration of vacancies which in turn have been increased by plastic deformation or quenching.

From Manjoine's measurements on soft iron at different strain rates, and from the known diffusion coefficient for N in  $\alpha$ -iron, Cottrell obtained the minimum rate of diffusion for repeated yielding in  $\alpha$ -iron in terms of the strain rate. From the difference in atomic concentration of the solute in Al and in  $\alpha$ -iron, and from

other differences in the two systems, he estimated that the Portevin-Le Chatelier effect should appear in Al alloys when

$$D \approx 10^{-10} \epsilon \text{ cm}^2/\text{sec.} \quad (1)$$

According to Seitz (Advances in Physics 1, 43 (1952)), plastic deformation should create vacancies, the concentration of vacancies so created being

$$c \approx 10^{-4} \epsilon. \quad (2)$$

The diffusion coefficient,  $D$ , for the motion of substitutional atoms in the presence of vacancies should be given by

$$D = ca^2 \nu \exp(-J/RT) \quad (3)$$

where  $a$  is the interatomic space,  $\nu$  is the frequency of atomic vibration and  $J$  is the activation energy to move a vacancy. Putting in the value  $a^2 = 10^{-15} \text{ cm}^2$ ,  $\nu = 10^{13} \text{ secs}^{-1}$ , and assuming  $J$  is one-third the activation energy of diffusion for Cu in Al, namely 10,000 Cal/mole, Cottrell obtains

$$D \approx 10^{-13} \epsilon \text{ cm}^2/\text{sec.} \quad (3a)$$

Comparing equations (1) and (3a) with various experimental data, Cottrell obtains agreement within a factor of ten for the strain at which the serrations begin. Considering all the approximations made, he feels that this degree of agreement is sufficient to lend support to the above explanation of the effect.

A more detailed account of this theory, together with a description of other metallurgical research at Birmingham can be found in Technical Report ONRL-53-53, available from the Technical Publications Office, Office of Naval Research, Code 740, Washington 25, D.C.

#### HISTOCHEMISTRY OF XANTHINE OXIDASE

G.H. Bourne of the London Hospital Medical College, in studying xanthine oxidase, the enzyme responsible for the oxidation to uric acid of the xanthine and hypoxanthine derived from the guanine and adenine of nucleic acid, has shown by biochemical means that the liver, kidney, spleen and intestine possess the highest activity

of this enzyme. Xanthine oxidase has not previously been localized histochemically, but the fact that it is a dehydrogenase suggested that it might be demonstrated by the use of neotetrazolium in the way that succinic dehydrogenase has been localized.

Frozen sections and thin slices of fresh un-fixed tissues were incubated in an atmosphere of nitrogen for one hour in a mixture containing pyrophosphate buffer, xanthine and neotetrazolium. The neotetrazolium was reduced to an insoluble diformazan pigment by the hydrogen liberated by the xanthine oxidase from the xanthine, and was precipitated at the site of enzyme activity.

In all tissues the nuclei were found to be completely negative, and in the gut the brush borders were negative. Cells in the red pulp of the spleen (which appeared to be reticular cells) gave a positive reaction. In the liver the peripheral cells of the lobules were more active than the central cells. In the kidney the greatest reaction was given by the cells of the proximal convoluted tubules and the descending limbs of Henle's loops. A less intense reaction was given by the other tubules. A slight reaction was given by the glomeruli. In the intestine all the epithelial cells of the villi were strongly positive, those of the crypts less so. The positive reaction consisted in each case of a diffuse red background (regions of low activity) and a bright blue granular reaction (regions of high activity). Both these reactions were distributed uniformly through the cell.

#### STUDIES ON THE CIRCULATION OF AUTOGRAFTS AND HOMOGRAFTS OF RABBIT SKINS

R.J. Scothorne and I. McGregor of the University of Glasgow report that the revascularization of full-thickness, fitted autografts and homografts on ear skin in rabbits has been studied by repeated intravenous injections of 4 per cent bromo-phenol blue. Skin with intact circulation is immediately colored blue following injection; skin grafts prior to vascularization are not immediately colored. Using this method it has been established that both autografts and homografts are fully revascularized by about four days following the placing of the graft. This is contrary to the findings of Ham (J. Bone and Joint Surgery, 34, 1952) and of Conway et al (Plastic and Reconstr. Surg., 9, 1952) who maintain that homografts of skin never

become vascularized at all. At some variable time after the re-establishment of the circulation, homografts are destroyed. Failure of the circulation, indicated by failure of bromo-phenol blue to enter the graft, is noted at the same time as histological signs of epidermal destruction. Since skin grafts can and do survive for periods of up to four days prior to revascularization, the investigators claim that it is unlikely that the destruction of homograft epithelium is due to this simultaneous vascular breakdown.

#### SOME MODIFICATIONS OF SILVER-SILVER CHLORIDE ELECTRODES

D.W. Kennard of the Pharmacological Laboratory, University College, London, has devised an improved form of silver-silver chloride electrode which has some advantages over the usual bare-wire type. Ligature silk woven in a hollow cylindrical form has been used to make the electrode.

A silver wire is sealed into a glass tube by polythene, which is fused on to the terminal portion of the tube. The wire is then threaded through the appropriate size of ligature silk, so that the latter projects beyond the end of the wire. The coated wire may now be bent into a hook or any shape required, and chlorided in the usual way. To keep the sleeve moist when in air, a small piece of cotton is placed around the base of the wire and soaked in saline. When stored it is kept immersed in saline.

The electrode has several advantages over bare chlorided wire. It maintains good fluid contact between any structure and the electrode, and the projecting end of sleeving can be used for contacting delicate surfaces. It helps to keep structures in contact with it moist, while the chloride film is protected from drying and the action of light.

Electrically it is more stable and much less polarizable than bare wire. The current passes between a nerve and bare wire, largely at the point of contact between the two, while a much larger area of the wire passes current when covered with the sleeve. This may be seen by examining the area of discoloration after passing relatively heavy currents.

Sleeving of this kind may also be used to improve the standard silver wire in agar electrode. The action of light may be reduced by using wire coils threaded with a sleeve. The emerging wick may be made rigid and any shape by employing a glass fiber covered with silk.

TECHNICAL REPORTS OF ONRL

The following reports have been forwarded to ONR, Washington, since the last issue of ESN. Copies may be obtained from the Technical Publications Office, Code 740, Office of Naval Research, Washington 25, D.C.

- ONRL-27-53 "Physiology of Athletics" by J.L. Nickerson
- ONRL-28-53 "Experiments on Blood Flow in the Liver Using A Standardized Thermocouple" by J.L. Nickerson
- ONRL-33-53 "The Application of Ultrasonic Techniques to the Study of Rotational Isomerism" by G.J. Szasz
- ONRL-37-53 "The Oxidation of Carbon Monoxide" by G.J. Szasz
- ONRL-38-53 "Physiological Research in Belfast and Dublin" by J.L. Nickerson
- ONRL-39-53 "The Elliott-N.R.D.C. Computer 401 Mark I" by R.R. Weber
- ONRL-43-53 "Notes on Physical Chemistry Research in Copenhagen" by R.W. Mooney
- ONRL-45-53 "Infrared Absorption Spectra and Configurations of Substituted Cyclohexanes" by G.J. Szasz
- ONRL-46-53 "British Electron Linear Accelerators in Commercial Production" by J.K. Beling
- ONRL-47-53 "Ministry of Supply Annual Counter Meeting, London, January 1953" by J.K. Beling and S.F. Singer
- ONRL-50-53 "Research of Semiconductors and the Theory of Alloys in Paris" by J.R. Reitz
- ONRL-51-53 "An Optical Image Intensifier" by W.L. Hyde
- ONRL-52-53 "Recent Meetings of the Ergonomics Research Society" by C.H. Graham
- ONRL-54-53 "Research on Luminescence and Related Solid State Phenomena in Berlin" by G.J. Szasz
- ONRL-55-53 "The Manchester Electronic Computer" by R.R. Weber
- ONRL-57-53 "Radiotherapeutic Research Unit, Hammersmith Hospital, London" by J.L. Tullis
- ONRL-59-53 "The Shape and Thermal Transitions of Macromolecules" by G.J. Szasz

ONRL-60-53 "The Radiobiological Unit of the Christie  
Hospital and Holt Radium Institute, Manchester"  
by J.L. Tullis

PERSONAL NEWS ITEMS

Professor R. Hilsch, Director of the Physical Institute, Erlangen, has accepted the invitation to the chair in physics at the First Physical Institute, Göttingen.

Dr. K. Lücke, who has been active in the study of the plastic deformation of metals at Göttingen University, has accepted an invitation to spend one year in research at Brown University.

Sir Lawrence Bragg, F.R.S. has accepted an appointment as Fullerian Professor of Chemistry, Resident Professor, and Director of the research laboratories at the Royal Institution. He will take up the duties of the Fullerian professorship immediately and the laboratory and resident duties on January 1, 1954. Sir Lawrence Bragg has been Cavendish Professor of Experimental Physics, Cambridge University, since 1938.

FORTHCOMING EVENTS

The Fifth Meeting of the International Committee of Electrochemical Thermodynamics and Kinetics will be held in Stockholm on 25 - 28 July. Details concerning the organization and program of the meeting are obtainable from the Secretary General of the Committee, Professor M. Pourbaix, University of Brussels.

Prepared by the Scientific Staff  
Submitted by Dr. S.R. Aspinall  
Deputy Scientific Director

  
PHILIP D. LOHMANN  
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Assistant Naval Attache for Research