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EUROPEAN SCIENTIFIC NOTES

Number No. 7 - 18,

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ELECTRON DISTRIBUTIONS IN IONIC CRYSTALS

Professor H. Witte (Darmstadt) has determined electron distributions in NaCl and LiF by means of a Fourier analysis of X-ray diffraction intensities from a large number of crystallographic planes in single crystals of these compounds. If each crystallographic plane, (hkl), diffracts X-rays with a certain intensity, $|F(hkl)|^2$, then as is well known, the electron density at a point (x, y, z) in the unit cell is given by

$$\rho(x, y, z) = \frac{1}{a_0^3} \sum_{h=-\infty}^{\infty} \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} F(hkl) \cos \frac{2\pi}{a_0} (hx + ky + lz),$$

where a_0 is the lattice parameter. Mo K_{α} radiation was used in the present study, the advantage of this short wavelength radiation being that it allows a large number of reflections. The X radiation from a Mo target was first monochromatized by reflection from a crystal (single reflection), and then was diffracted from the ionic crystal under study. Witte has measured the diffracted intensity from about 100 crystallographic planes in each crystal by means of an ionization chamber. The computation of the electron density was carried out only for points (x, y, 0) in crystal planes containing the centers of gravity of the ions; even so the procedure

has been a laborious one, and has taken about one year for each compound.

The results of the investigation show that sodium chloride is a highly ionic crystal with a region of zero charge density between the sodium and chloride ions. The density contours in a plane containing the centers of gravity of the ions are shown in Fig. 1a; 14.5 to 15 of the electrons in Cl^- ion are spherically symmetric. The results for lithium fluoride show that this compound is also highly ionic, although the minimum charge density between ions is about 0.2 electrons per Å^3 . The density contours for this crystal are shown in Fig. 1b. The density distribution in LiF

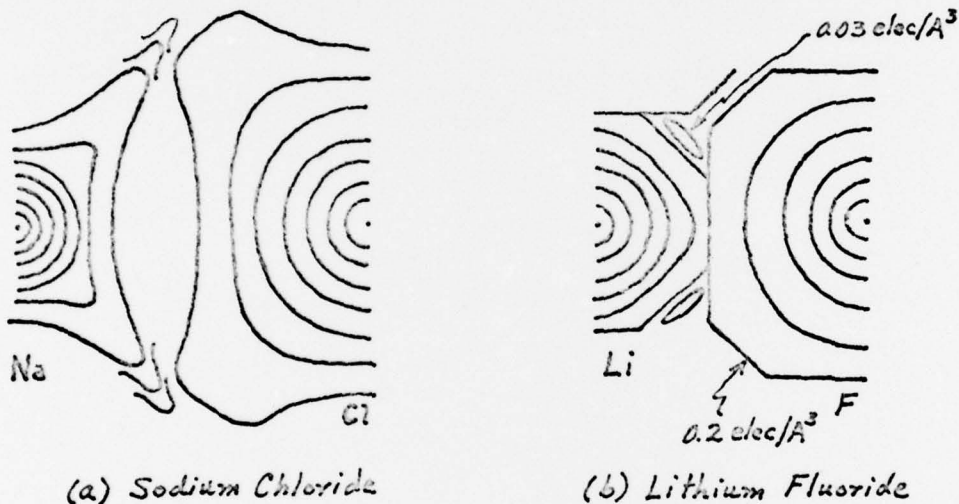


Fig. 1. Charge Density Contours in Two Ionic Crystals
(In a plane containing centers
of gravity of the ions)

shows practically no homopolar binding; thus the results disagree with the calculation of Ewing and Seitz (Phys. Rev. 50, 760 (1936)).

A review of the solid state physics research program at the Technical University, Darmstadt, is given in Technical Report CNRL-109-53.

INTERNATIONAL MEETING ON MOLECULAR SPECTROSCOPY

An informal international Conference on Molecular Spectroscopy, held in Paris, 29 June - 3 July, afforded a valuable survey of the activities of the various European spectroscopy laboratories. Over one hundred papers were presented, including virtually all branches of spectroscopy. The meeting was predominantly experimental, with most of the papers devoted to infrared and ultraviolet spectroscopy. Preliminary plans for this meeting were made at the previous one at Basel two years ago (cf. ESN 5, 172 (1951)); it appears likely that these meetings will become a regular biennial feature of the European scientific scene. The persons primarily responsible for the success of these meetings are H. W. Thompson (Oxford), J. Lecomte (Paris), and R. Mecke (Freiburg).

Technical Report ONRL-107-53, summarizing some of the papers, is available from the Technical Publications Office, Code 740, Office of Naval Research, Washington 25, D. C. It is planned to publish short (French) versions of all the papers presented, in the *J. de Phys.* (Paris).

Spectroscopy at High Pressures

Recent results on infrared and ultraviolet absorption spectra of gases at high pressures were discussed by workers from Amsterdam, Toronto and Paris. Fahrenfort, Kluiver and Babeliowsky (Amsterdam) observed the pressure-induced infrared absorption spectrum of carbon dioxide in the region of $700 - 7300 \text{ cm}^{-1}$ at pressures up to 125 atmospheres and temperatures up to 135°C . A number of new absorption bands were observed; these were identified and said to fall into two groups, one corresponding to the allowed very weak transitions of the unperturbed molecule, the other being the forbidden transitions activated by the pressure-induced perturbations (cf. ESN, 7, 144 (1953)). Professor H. L. Welsh (Toronto) cast considerable doubt on the validity of these Amsterdam results. He pointed out that carbon dioxide at high pressures is an extremely good solvent of oil and that in his opinion most, if not all, of the new bands reported may be due to impurities.

Professor Welsh presented his own recent results on infrared absorption spectra of hydrogen at pressures up to 1500 atmospheres, which have led to the discovery of some new effects. The anomalously high rate of increase of the absorption coefficient at these pressures is ascribed to the finite volume of the molecules. The Q branch of the fundamental absorption undergoes increasing splitting with increasing density of the perturbing gas.

The effect of high-pressure inert gas on the rotational structure of the fundamental HCl band was reported from the laboratory of Dr. B. Vodar (Bellevue). The rotational structure is retained in argon to a much higher density than in nitrogen; in addition, with nitrogen a pressure-induced Q branch appears near the center of the band whose intensity increases rapidly with nitrogen pressure. An unsuccessful search was made for a comparable Q branch in carbon monoxide. The shape of the perturbed HCl band is similar to that which has been observed for this molecule in certain solvents.

The Intramolecular Inversion of Triethylamine

The dielectric absorption of a number of amines in dilute Nujol solutions, and for some in the pure liquid state, was discussed by Dr. Mansel Davies and Dr. A. H. Price (Aberystwyth). Their experiments covered the frequency range 10^4 to 10^8 cps. In the case of triethylamine they apparently succeeded in observing the inversion frequency with the characteristic relaxation time of 0.44×10^{-8} sec. The dielectric loss curve observed for this substance does not conform to the Debye relations for dipole relaxation but the results fit very well the contour deduced by Fröhlich for a resonance absorption. Investigating the concentration dependence of this resonance absorption the product of the characteristic time and the concentration was found to be constant; this is apparently analogous to the fact that in ammonia the product of the pressure and the relaxation time of the inversion is a constant quantity. A curious consequence of this analogy is the indication that only collisions between triethylamine molecules can activate the inversion.

Spectroscopic Evidence for an Isomeric Form of Hydrogen Peroxide

As is well known, condensation of the products of an electric discharge through water vapor at liquid air temperatures yields a glassy material. This undergoes a transformation around -115°C to give a hydrogen peroxide solution of about 50% strength. The possibility that an isomeric form of hydrogen peroxide, H_2O_2 , is involved has been repeatedly discussed with no definite conclusions being reached. Professor P. A. Giguere (Quebec, Canada) has observed the infrared absorption spectrum of the glassy material and reports a new band of medium strength at 1300 cm^{-1} . Using heavy water, the same new band has again been observed, while all other bands undergo an isotopic shift. Thus, this new band is not due to a hydrogen vibration and may well correspond to the semi-polar O-O band involved in the hypothetical, isomeric hydrogen peroxide structure.

Analogy Between Radio Frequency Transitions and Raman Effect

Professor A. Kastler (Paris) drew attention to a significant analogy between radio frequency transitions involving two quanta and the Raman effect. He referred to the experimental results of Grabner and Hughes (Phys. Rev. 82, 596 (1951)) who observed transitions between the electric-quadrupole hyperfine-structure levels of Rh^{85}F by means of the molecular-beam electric-resonance method. They reported a transition produced by the simultaneous action of two electro-magnetic quanta such that the sum, $h\nu_1 + h\nu_2 = \Delta E$. Professor Kastler pointed out that such a transition is formally analogous to those which in the optical spectra give rise to the Raman effect, and that this analogy may be exploited to predict selection rules and also rules for polarization phenomena. He suggested that a search for transitions in the radio frequency range involving differences between quanta such that $h\nu_1 - h\nu_2 = \Delta E$ would be of interest.

The Structure of Methyl Nitrite

The Raman spectrum of methyl nitrite was investigated at $+20^{\circ}$ and -80°C by Dr. J. Wagner (Graz). Only two of the Raman lines are temperature dependent, and Wagner believes that these changes are due to the progressive breakage of the internal hydrogen bond in the cis form without, however, appreciable amounts of the trans form being formed. These results are thus at variance with those of Tarte (J. Chem. Phys. 20, 1569 (1952)) who interpreted infrared and ultraviolet spectra data in terms of rotational isomerism between the cis and trans forms.

DISCUSSION ON RECENT PROGRESS IN THE STUDY OF MOLECULAR STRUCTURES

The annual meeting of the Societe de Chimie Physique, Paris, 6 - 10 July, was devoted to "Recent Progress in Molecular Structure Studies". The meeting was organized on somewhat novel lines; each section was introduced by a survey lecture which was available in preprint form, and this was followed by an open discussion. While most of the physical methods of current importance in the elucidation of molecular structure problems were reviewed, relatively few new results were presented; a significant proportion of these came from the Oxford electron diffraction group under L. E. Sutton and some of these are briefly described below.

The contributions of the Russian participants on Raman intensities (Dr. P. P. Shorygin) and on paramagnetic resonance (Dr. P. M. Kozyrev) were interesting, but in regard to experimental results, did not go beyond work they have previously published.

Theories of the Chemical Bond

The discussion on theories of the chemical bond was devoted largely to two aspects of the subject. (a) Numerous examples were cited to show the importance of unshared electron pairs in stereochemistry. The unshared electron pairs are generally similar to shared ones in their stereochemical consequences. (b) The importance of d and f orbitals in chemical binding was the other problem of major interest. L. Orgel (Oxford) extended the overlap suggestions of Mulliken and of Maccoll from s-p orbitals to include also d orbitals. He stated that the relative strength of the sp^3d^2 octahedral orbitals to the sp^3 tetrahedral ones is 2.5 to 2, rather than 3 to 2 as predicted by Pauling. He also emphasized the importance of atomic size in orbital overlap considerations. Large d orbitals can form p bonds with small orbitals; a good example of this is provided by a comparison of trimethyl amine with its silicon analogue: $N(CH_3)_3$ is pyramidal and a strong base, while $N(SiH_3)_3$ is planar and nonbasic.

The Structure of Ferrocene

The following parameters were obtained by Seibold and Orgel in an electron diffraction study of ferrocene vapor (dicyclopentadienyl-iron)

Fe - C distance	$2.04 \pm 0.01A$
C - C distance within the rings	$1.43 \pm 0.01A$
C - H distance	1.09 ± 0.02

The equilibrium configuration is one in which the parallel rings are staggered with respect to one another, as would be expected if hydrogen repulsions are operative. It is interesting that the C-C distance reported here is considerably longer than that in benzene. Dr. G. Fimentel (Berkeley) pointed out that recent infrared and Raman spectra of this

substance indicate that the two five-membered rings are completely independent as regards any coupling between symmetrical and antisymmetrical ring vibrations.

CONFERENCE ON THE THEORY OF THE PLASTIC DEFORMATION OF METALS

At a recent informal Conference on the Theory of the Plastic Deformation of Metals held at Bristol University, papers were presented on various aspects of plastic deformation including creep, tensile and shear behavior of monocrystals, and the fatigue of metals. In the following, two of the most interesting contributions are summarized. A more complete description of the meeting is given in Technical Report CNRL-108-53.

The Rate of Fatigue Crack Growth

Dr. A. K. Head (Melbourne and Bristol) has made a theoretical treatment on the rate of growth of a fatigue crack. As a model he chose a crack in an elastic medium with an alternating (tensile and compressive) stress perpendicular to it. In such a case a stress concentration exists at the ends of the crack and therefore plastic deformation occurs to dissipate the intensified stress. This process cannot continue since the material work-hardens, and thus the crack grows into the material by extension of its ends. The entire process repeats itself during succeeding cycles, and the crack continues to grow.

To simplify the mathematical treatment, the elastic medium was replaced by a set of interacting mechanical elements; it was further assumed that the element at the head of the crack had an elastic-plastic behavior, and that the amount of work-hardening per cycle is low. The result of Head's derivation is the following equation:

$$dL/dn = KL^{3/2},$$

$$K = (F/12E) s^3 (s_1 - s_0)^{-1} (s_0 - s)^{-2} \alpha_0^{-1/2},$$

where L = length of crack,
 n = number of cycles,
 F = plastic modulus or rate of work-hardening,
 E = elastic modulus,
 S = applied stress,
 S₁ = tensile strength or fracture stress,
 S₀ = yield stress,
 α₀ = width of the plastic region around the tip
 of the crack.

This relationship leads to the following equation for the crack length as a function of number of cycles:

$$L^{-1/2} = (K/2) (N - n)$$

where N = number of cycles for complete fracture.

Using the data for fatigue crack growth from Bennett (Proc. ASTM 46, 693 (1946)), DeForest (Journal Applied Mechanics) and Moore (1928), Head has obtained straight lines in plots of L^{-1/2} versus the number of cycles. Extrapolation of Bennett's data to zero number of cycles gives an initial crack size in the material of 5 x 10⁻⁴ inches. Further, analysis indicates that there is no stress at which an existing crack will not grow.

The analysis does not take into account the contributions to crack growth made by the joining up of numerous small cracks, as is known to occur in certain cases. In addition, it may be criticized that the theory is contrary to the observation that fatigue cracks do, on occasion, cease to grow with successive cycles. The details of Head's work will be published in Philosophical Magazine.

Strain Hardening of Aluminum Monocrystals

Professor A. H. Cottrell (Birmingham) described some work done in collaboration with Dr. H. W. Paxton (formerly Birmingham, now Carnegie Institute of Technology) on the effect of torsion on the tensile behavior of monocrystals of pure aluminum. The results of this experiment clearly show the importance of dislocations on two sets of slip planes interacting with each other.

Briefly, the experiment consists of pulling a single crystal (3 mm diam., several cm long) in tension to about 1% extension, removing the tensile load, producing a permanent deformation in torsion of three minutes of arc per centimeter of length, and finally continuing the tensile test. It is observed that the twist produces considerable work-hardening in the crystal (see Fig. 1). The effect of the twist on the

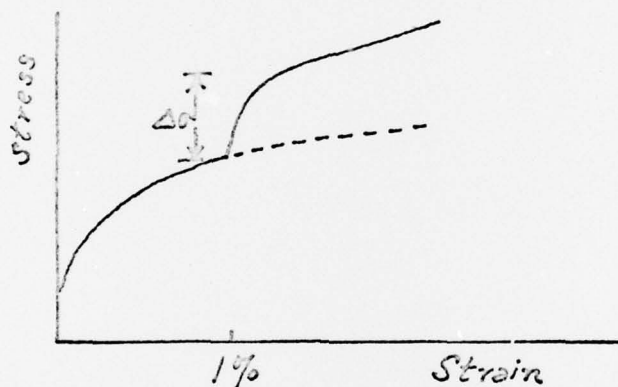


Fig. 1. Effect of torsional deformation on the stress-strain curve of an aluminum monocrystal

stress-strain curve is much larger than that which would be expected from the surface strain resulting

from the twist (surface strain $\approx 3 \times 10^{-4}$ for this case). Plastic twisting is ideal for putting dislocations into the crystal, and the slip systems in torsion are oblique to those in tensile slip. The work hardening resulting from the twist is thus explained by the presence of dislocations, produced on oblique planes as a result of torsion, which then interact with and restrain the movement of dislocations in the subsequently operative tensile slip planes.

Cottrell has calculated the stress ($\Delta\sigma$) needed to drive a tensile dislocation through an array of oblique dislocations with a mean spacing, ℓ . To cut through one row of the latter, a portion of length, ℓ , of the tensile dislocation has to move a distance of order of the Burgers vector b , and the work done by the applied stress on it is $(\Delta\sigma)\ell b^2$ (b is taken as one atomic spacing for the average dislocation). The result of a dislocation cutting through another is to produce a dislocation jog one atomic spacing long; the energy of this jog is aGb^3 , where G is the shear modulus and a is a numerical constant of the order of unity (see A. H. Cottrell, *Dislocations and Plastic Flow in Crystals*, Oxford (1953), p. 174). Thus equating the two energy terms:

$$(\Delta\sigma)b^2\ell = aGb^3,$$

or

$$\Delta\sigma = aGb/\ell. \quad (1)$$

Since the dislocations produced by torsion are fairly uniformly distributed in the tensile slip plane, equation (1) can be written as

$$\Delta\sigma = aG\rho^{1/2}b, \quad (2)$$

where $\rho = \ell^{-2}$ is the density of intersecting dislocations. Thus $\Delta\sigma$ can be evaluated from the density, ρ , of dislocations produced as a result of torsion, which in turn can be found by consideration of a Burgers circuit. A Burgers circuit is defined as an atom-to-atom circuit traversing only "good" regions of

the crystal; in a perfect crystal the Burgers circuit is closed, but in an imperfect crystal the circuit misses complete closure by the total Burgers vector enclosed in the circuit. See W. Shockley et al, Imperfections in Nearly Perfect Crystals, Wiley, New York (1952), Chap. II. Cottrell considers a square circuit of length d on a side in a plane perpendicular to the motion of twist; if θ is the plastic twist per unit length, the circuit misses being closed by θd^2 . The total number of dislocations piercing the circuit is ρd^2 and thus the total Burgers vector piercing the circuit is $\rho d^2 b$. Therefore,

$$\rho = \theta/b$$

$$\text{and } \Delta\sigma = \alpha G(\theta b)^{\frac{1}{2}} . \quad (3)$$

Putting $\alpha = 1$, inserting the appropriate G and b values for aluminum in equation (3), and taking $\theta = 3'$ arc/cm, Cottrell computes $\Delta\sigma = 20$ grams per square millimeter, which is in excellent agreement with the experimentally observed increase in stress shown schematically in Fig. 1.

SECOND SYMPOSIUM ON GASDYNAMICS OF THE INTERSTELLAR CLOUDS

A small symposium, sponsored by the International Union of Theoretical and Applied Mechanics (IUTAM) and the International Astronomical Union (IAU), was held at Cambridge, England, 6 - 11 July, for the purpose of discussing problems in the gas-dynamics of interstellar clouds. The subjects treated included turbulence of interstellar gas, various types of observed discontinuities, the nature of the spiral arms of nebulae, collision phenomena, accretion, and the constitution of interstellar clouds. Publication of the proceedings is planned under the editorship of J. M. Burgers (Delft), H. C. van de Hulst (Leiden), and J. H. Oort (Leiden). The symposium is surveyed in Technical Report CNRL-101-53.

Cosmical Turbulence

Observations indicate that interstellar clouds are in a state of random motion which has been termed turbulence. This turbulence is very much different in nature from turbulence of the terrestrial variety and is characterized by large values of the turbulent Mach number, i.e., the ratio of the mean turbulent velocity to the mean velocity of sound. The characteristics of this type of turbulence were brought out by the contributions of M. J. Lighthill (Manchester), G. K. Batchelor (Cambridge), and J. M. Burgers (Delft). Compressible effects must be of major importance in comparison with shear effects, the turbulent pressure and energy must be large compared with the thermodynamic pressure and energy, and the major portion of the energy dissipation by viscosity and heat conduction must occur directly rather than by means of a cascading process such as is present in the turbulence of an incompressible fluid.

Magnetohydrodynamic effects are expected to be important in cosmical turbulence because of the high conductivity of the interstellar gas. According to Batchelor, magnetic energy is produced as a result of the stretching of the magnetic lines of force by the small scale motion, and equipartition should be expected between the magnetic energy and the small scale turbulent energy. On the other hand, Fermi has suggested that equipartition should be between the magnetic energy and the total turbulent energy. One difficulty in the resolution of this controversy is the present lack of understanding as to the nature of high Mach number turbulence.

Zanstra's Equivalent Equation of State

One of the interesting contributions in the field of discontinuities was that of H. Zanstra (Amsterdam), who considered steady-state energy balance of a strongly irradiated gas corresponding to conditions characteristic of planetary nebulae. The gas is heated and ionized by the incident radiation and is considered able to lose energy through the excitation of a low-level excited state (N II or O III). This loss of energy through the low-level metastable state is much more effective at high densities, and with a given exterior radiation temperature and a given pressure, more than one steady state solution for the density and electron temperature can exist. With suitable simplifying assumptions Zanstra has plotted pressure versus volume for such a gas, with exterior radiation temperature as a parameter, and finds that the resulting curves appear as an inverted van der Waal's gas. If the exterior temperature is low the resultant curve is without inflection and no condensation may be effected. However, if the exterior temperature is high, then three solutions for the density are possible in certain pressure ranges. As with a van der Waal's gas, the existence of a phase discontinuity is deduced. Although others have studied such discontinuities for interstellar material, Zanstra's contribution presents a conceptual simplification which is very welcome.

A New Spiral Arm in Our Galaxy

J. H. Oort (Leiden) reported recent findings with the radio telescope (21 cm H radiation), principally the indication of the presence of a second spiral arm outside the one in which our sun is located; this is in addition to the outer arm previously found.

Collisions of Interstellar Clouds

F. D. Kahn (Manchester) showed that the interpenetrating flow after the collision of two completely ionized gas clouds of very large mean free path must be unstable. With reasonable assumed values for the

density of the gas, the time constant for the instability of the electron stream should be of the order of 10^{-6} sec, and that for the proton stream of the order of 1 sec. The conclusion is that such colliding gas clouds would not be able to interpenetrate.

In another contribution Kahn discussed a mechanism for the radiation of thermal energy produced by the collision of interstellar clouds. He suggests that small ice splinters with linear dimensions of about 10^{-6} cm or less are present in the gas and are responsible for the efficient dissipation, by radiation, of heat-energy absorbed from the gas. As one possible mechanism for this radiation, Kahn suggests that the splinters are cylinders of polar ice and are cool enough to be covered by a layer of H atoms; each splinter would have positive and negative regions on which would be found ions (H^+ and H^-) which can cause radiation of energy through plasma oscillations on the cylinder surface. For this process the temperature of the ice splinter must be below 8° K.

TECHNICAL REPORTS OF ONRL

- ONRL-76-53 "The German Society for Applied Mathematics and Mechanics" by W. D. Hayes
- ONRL-91-53 "Notes on the Fifth International Congress of Otorhinolaryngology" by J. L. Tullis
- ONRL-96-53 "Metallurgy in Italy" by E. Epremian
- ONRL-97-53 "The Activities of Some Psychologists in Belgium, The Netherlands, and Spain" by C. H. Graham
- ONRL-98-53 "Notes on the Second European Congress of Allergology" by J. L. Tullis

PERSONAL NEWS ITEM

Professor F. A. Paneth of the University of Durham has accepted an invitation to join the Max Planck Institute of Chemistry in Mainz, Germany. He will assume his new position this autumn and will be succeeded at Durham by Dr. G. E. Coates of Bristol University.

FORTHCOMING EVENTS

At the recent Cosmic Ray Congress held at Bagneres, France, Professor P.M.S. Blackett announced the decision of the IUPAP Subcommittee on Cosmic Radiation to hold the next meeting in September 1955. Mexico City appears as a likely possibility for the location of the Congress, since an invitation has been extended by Professor M. S. Vallarta. The subject matter of the forthcoming Congress will be devoted to the primary radiation, extensive air showers, time variations and correlations, and theories of the origin of cosmic rays.

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