the physics of LIQUIDS

. . . a conference report

By Joseph L. Hunter and Edward F. Carome

Under the sponsorship of the National Science Foundation, the Physics Department of John Carroll University played host to about sixty researchers in the field of the physics of liquids at a four-day conference from June 1 through June 4, 1964. Although the meeting was built around a nucleus of prepared talks, several of the participants preferred the more informal approach of participating from the floor in all of the talks. Among the latter were Daniele Sette of the University of Rome, Henry S. Frank of the University of Pittsburgh, Martin Greenspan of the National Bureau of Standards, and Robert T. Beyer of Brown University. As a matter of fact, most of those delivering talks preferred to think of themselves as discussion leaders rather than lecturers.

Successive half-day sessions were devoted to the following: viscosity and viscoelasticity; x-ray and neutron diffraction by liquids; nuclear magnetic resonance effects and positron annihilation in liquids; dielectric and ultrasonic relaxation in liquids; and liquid chemistry. A full day was devoted to general liquid theory.

The first day was given over substantially to general liquid theory, handled by Peter Gray, of The University, Newcastle-on-Tyne, and by Herbert S. Green, of the University of Adelaide. Their task was to give to the assembled group, composed mainly of experimentalists, some notion of the newest approach and emphasis in liquid theory. Gray opened the seminar with the statement that the statistical mechanical theory of liquids may be conveniently divided into two parts, equilibrium and nonequilibrium theory, and made the point that the two are, at the present time, qualitatively different. The equilibrium theory is formally exact, and well-defined mathematical approximations are introduced to obtain numerical results. On the other hand, the nonequilibrium theory requires approximations of a physical or intuitive nature.

For the equilibrium case, Gray described the fundamental problem: the expression of thermodynamic quantities in terms of the molecular potential and the radial distribution function. The radial distribution function involves a power series in the density in which the coefficients are irreducible cluster integrals (or diagrams). Representation of the density requires a summation over the various types of diagrams. Different closed integral equations are produced, depending on the approximation used in the summation, the most prominent being the Yvon-Born-Green, the hyper-netted chain, and the Percus-Yevick. Agreement is generally good at low densities, but becomes progressively worse at higher densities.

In nonequilibrium theory, Gray discussed formulae for the viscosity, thermal conductivity, and diffusion coefficients. These are obtained again in terms of the pair potential and radial distribution functions by solving the kinetic equations to first order in the velocity, temperature, and concentration gradients. The agreement of the numerical values so obtained with experiment is reasonably good and strongly correlated with that of the thermodynamic functions. An important outcome of these calculations is that the theory is entirely unequivocal as to the existence of a bulk viscosity; in the case of liquid argon, for example, its calculated value varies between one and three times the shear viscosity at different temperatures and densities.

In his talk, Green chose to describe in some detail a modern problem in the statistical mechanics of equilibrium processes, and one in nonequilibrium processes. For equilibrium processes, he chose a modification of the Monte Carlo method as an illustration. A set of M ($M \sim 25$) particles is started from a random configuration in a box with periodic boundary conditions. Each particle is visited in turn; the particle is either left in its position P or displaced to a randomly chosen neighboring point P', according to whether a ran-

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Joseph L. Hunter of John Carroll University was the director and Edward F. Carome of John Carroll University and Ernest Yeager of Western Reserve University were co-directors of the conference reported here. Included also in the planning were William Cramer of the Office of Naval Research and Theodore A. Litovitz of Catholic University.

dom number between zero and one exceeds or is less than a function of the potential energy of the two particles (which function may also vary between zero and one). Favored distributions result from this process, and Green described the results of his work in applying the results to electrolytes. Two intriguing results are that pairs of opposite charges predominate at temperatures below $10^4/K$, where K is the dielectric constant, and that electric waves may result from an initial nonequilibrium ensemble.

Green also described, in some detail, a method of dealing with irreversible processes in which the evaluation of the all-important autocorrelation functions is reduced to the solution of a (comparatively) well-known hierarchy of integral equations for the few-particle distributions. The method is based on the formalisms of Kubo, Mori, M. S. Green, and H. S. Green. Heretofore, such equations were, practically speaking, unsolvable. However, with the help of advanced computational techniques and improved approximations of the hyper-netted chain type, there is now hope that they will finally yield. Green estimated that, within the next few years, they will allow transport coefficients for liquids to be evaluated with an accuracy similar to that obtained for dynamic variables.

In order to lighten the load on the various participants during the first day of the conference, several other talks were interspersed between portions of the theoretical ones presented by Gray and Green. In one of these, Carome discussed the results of several experiments on laser-induced acoustic effects performed by his research group at John Carroll University. Intense plane-wave acoustic impulses have been generated in an optically absorbing liquid layer using the defocused beam from a Q-spoiled ruby laser. He indicated that such signals might be of use in studying relaxing liquids. The focused beam from a similar laser also has been used to generate wide-band ultrasonic and hypersonic waves in liquids and solids, and acoustic signals in excess of two kilomegacycles have been propagated and detected acoustically in various liquids. Though stimulated Brillouin scattering is probably the source of some of the observed signals, it appears that sources such as dielectric breakdown also are active.

Jacek Jarzynski of the American University described his ultrasonic measurements in the alloys of molten metals. Although there were many interesting points in regard to these experiments, perhaps the most interesting was the variation of volume viscosity, particularly its variation with the percentage of different alloy materials. Jarzynski gave results for potassium, sodium, silver, tin, and several other pure metals and combinations of these metals in alloys. A tin-silver alloy is a case in point. For large percentages of tin, the alloy manifests a large volume viscosity (a ratio of volume-to-shear viscosity of about five to one). However, as the percentage of silver is increased, the ratio of volume-to-shear viscosity falls rapidly, becoming less than one and approaching zero asymptotically, although Jarzynski did not actually reach the zero value experimentally.

The morning of the second day was devoted to viscosity and viscoelasticity, primarily (but not entirely) from the experimental approach of ultrasonic propagation. As was the case with Gray and Green in general liquid theory, this was a cooperative endeavor of Joseph L. Hunter of John Carroll University, Theodore A. Litovitz of the Catholic University, and John Lamb of the University of Glasgow. Hunter laid the groundwork for present-day theories of viscoelasticity, starting with the theory of the ideal liquid, with its single elastic constant. He then showed that this approach may first be generalized by the introduction of a second constant; if this is a viscous constant, one has the theory of viscous liquids; if it is a second elastic constant (the shear elasticity) one has solid elasticity theory. He discussed the partial generalizations possible, and arrived at the one unique to present-day viscoelastic theory, defining the various viscoelastic moduli. He also gave the historical background to the concept of the bulk viscosity. He concluded with a description of most recent measurements which enable the evaluation of the viscoelastic constants.

Litovitz used the viscoelastic constants, in particular the relaxational compressional modulus, to introduce a well-knit theory of viscosity in which free volume figures vary prominently. He indicated that the relaxational moduli and the free volume should be closely related, although this relation has not been evident until recently because of the comparatively little information on the moduli. Litovitz reviewed the best-regarded recent theories of viscosity, pointing out the strengths and weaknesses of each; he then showed that a theory which he proposed fits experimental values better than the existing theories. Very briefly this theory takes into account the facts that a molecule not only must have the strength to break a bond, but it must also have a space to go to if it is to succeed in breaking it.

Whereas Litovitz was primarily interested in the compressional modulus, Lamb's talk was devoted

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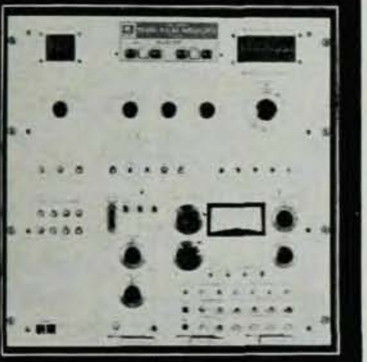

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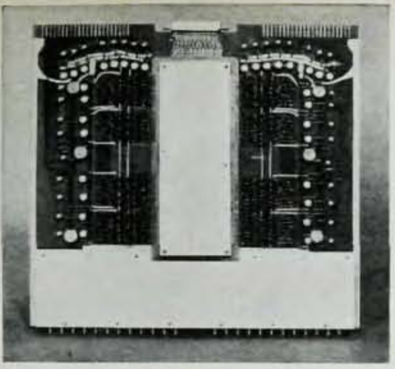

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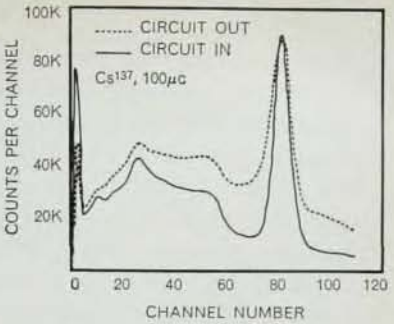

Cs^{137} spectra accumulated at high counting rates with and without pile-up rejection circuit.

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to measurement of the shear modulus. He described the values of this modulus obtained in a number of silicone liquids. These are mixtures of linear polysiloxanes of varying molecular weights. The shearing modulus is measured by reflecting the shear waves at a quartz-silicon interface. It is possible, though very difficult, to measure the phase, as well as the magnitude, of the shear reflection coefficient. Lamb was able to obtain the angle of the shear reflection coefficient up to a frequency of 70 megacycles, which was sufficient for the purpose of the experiment.

The afternoon of the second day was devoted to diffraction by liquids: George W. Brady of Bell Telephone Laboratories spoke on the diffraction of x rays and P. A. Eglestaff of the British Atomic Energy Research Establishment (Harwell) spoke on the diffraction of neutrons.

Brady discussed the fundamentals of the theory of large and small angle diffraction, and then described the major experimental techniques and the major difficulties involved in diffraction by liquids. He chose a very interesting representative analysis: the structure of $FeCl_3$ in acid and neutral solution. A striking finding was that the coordination was octahedral in neutral concentrated solution, whereas in acid the solute turned into a polymeric form of alternating tetrahedral and octahedral units. In addition to the geometry of the basic units, x-ray diffraction has also been found useful as a clue to clustering in the critical region. From this viewpoint, Brady discussed various forms of correlation found in the solution $C_7F_{16} - C_8H_{16}$.

Eglestaff discussed the scattering of slow neutrons in terms of the probability of exchanging energy between the neutrons and the system when a momentum transfer takes place. From this probability function a Fourier transform is obtained, part of which is connected with the neutron-scattering pattern. The behavior of the scattering gives qualitative information about the asymptotic behavior of the atoms; for instance, for one special condition it gives information about the way in which slow diffusion processes in normal liquids take place. Two extremes of behavior may be distinguished, which Eglestaff termed Lorentzian and Gaussian. The former is characterized by slowly fluctuating interactions with other atoms, and the latter by rapid fluctuations. These influence the line shape of the scattering distributions. Thus one gets quantitative information of the velocities of motion involved and the distances between collisions. Also, the scattering function may be related to other correlation functions. There is a case in which the scattering is proportional to the velocity-correlation function, and this provides a means of determining the number of degrees of freedom for particular modes of motion: e.g., modes leading to diffusion. In another instance, the scattering function can be related to the properties of sound-wave propagation in the system. In general, one must beware of adopting too naive a concept of the relation between neutron diffraction and liquid structure as such.

J. G. Powles of Queens College, University of London, began his talk on nuclear magnetic resonance in liquids by saying quite emphatically that he was mainly concerned with using nuclear-magnetic-resonance techniques in studying the rate and the nature of molecular motion in liquids, and not in elucidating the structure of the molecules themselves. He stated that nmr is an ideal tool for this, since the measured quantity, nuclear magnetization, is very sensitive to molecular motion but has quite a negligible reaction on molecular motion. Only a very restricted part of the molecular motion is "seen" by nmr. (Measurements in benzene have clearly demonstrated that the nuclear magnets see only the low-frequency Fourier components.) However, in spite of this, nmr is quite successful in evaluating correlation times of molecular motion. In iso-butyl bromide, correlation times varying with temperature over the range from 10^{-2} to 10^{-11} sec have been deduced, and are confirmed by the more direct measurement of dielectric relaxation, which depends on a closely related correlation function. Powles then described interesting findings by nmr with respect to the degree of difference between solid-liquid and liquid-vapor close to their critical points. Basically, these indicate that the microscopic difference between phases is not as marked as macroscopic properties would suggest.

Powles also mentioned the advantage of nmr because of the possibility of varying parameters such as pressure, temperature, and composition with relative ease because of the relatively remote contact between the sample and the measuring device.

Powles had been interrupted several times in his talk by those defending rival interpretations, or otherwise displeased by his forthright approach. Intransigent to the end, he concluded with the hope that data from nmr and other related methods would "save us from the present unhealthy and empirical recourse to the discussion of dubious concepts such as activation energy and microviscosity and so on". The ensuing discussion was noisy.

Leonard A. Roellig of Wayne State University

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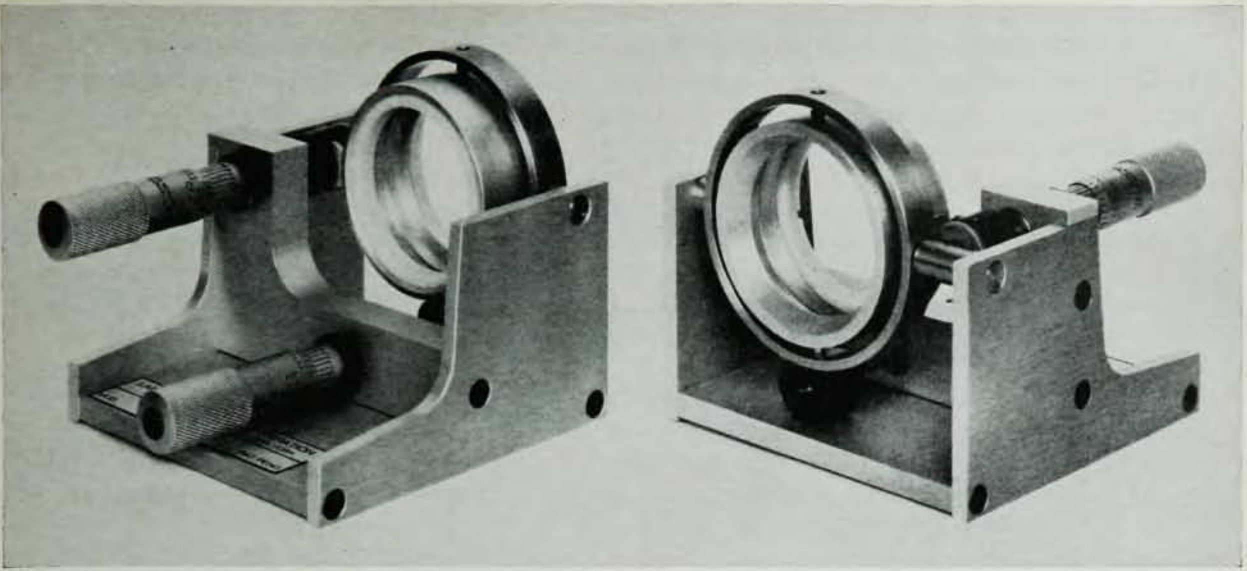

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See us at the 1965 13th Annual Physics Show—Statler Hilton Hotel, New York City—January 27-30, 1965. Booth 117 discussed the relation between positron annihilation and many properties of liquids which are of fundamental interest. Briefly, the characteristics of positrons are influenced by their environment. These characteristics include the lifetimes of the free positron, and singlet and triplet positronium; the rate of formation of positronium, and the rates of the two-gamma and three-gamma annihilation modes of positronium. All these are found to depend sensitively upon the physical state, molecular composition, pressure, temperature, and other parameters of a liquid. Roellig discussed the various experimental methods employed in positron annihilation, and gave experimental results in liquid metals, cryogenic liquids, and conventional liquids. He also described some very recent measurements of his own in superfluid helium and in teflon. He concluded by describing in general how positron annihilation would be employed in three important cases: the Fermi surfaces of liquid metals, solid-liquid phase changes, and microscopic density changes in fluids.

Robert Cole of Brown University, in his talk on dielectric polarization and relaxation in liquids, began with the following points which he considered important for a present-day understanding of dielectric measurements: (1) the approximate character of such quantities as polarizability and permanent dipole moments as molecular constants; (2) the approximate validity of the Lorentz field for nonpolar liquids; (3) the failure of the Lorentz field for polar liquids; (4) the need in some cases of considering quadrupole interaction fields and energies.

He mentioned that the Kirkwood theory of static dielectric constants, modified to treat induced moments, consistently has given good semiquantitative results when used to study local equilibrium correlations of a representative dipole and its neighbors. It accounts quite well for the large dielectric constants and temperature coefficients of HF, HCN, and the alcohols. He then described an extension of Kirkwood's equilibrium theory by Kubo, Glarum, and himself which relates dielectric relaxation to the time-dependent correlation function of a dipole with itself and its local environment. This theory indicates that one should not expect major differences between macroscopic and microscopic functions. Cole gave examples leading to single and multiple relaxation times. He also surveyed what he considered the most interesting recent developments in the field of dielectric relaxation. He compared the simple behavior of the aliphatic alcohols with the nonexponential relaxation functions of the glycols and the alkali halides. Finally he noted that broad relaxation spectra need not imply distributions of relaxation times; they may rather result from cooperative processes which are intrinsically nonexponential in time.

George McDuffie of Catholic University traced out the relations among the findings in the three fields of measurements which may be represented as ultrasonic, dielectric, and nuclear magnetic. In each case, information is gained with regard to relaxation processes in liquids. Static viscosity measurements serve to supplement these. If one restricts one's attention to associated liquids, certain similarities in the behavior of the static viscosity and the characteristic times for ultrasonic, dielectric, and nuclear magnetic processes become evident. As an example, the temperature dependence is similar and shows a non-Arrhenius behavior. Similarities can also be noted with respect to pressure dependence and effects of impurity molecules. Again, the activation enthalpy is nearly the same for all four processes. The dielectric relaxation time tends to be larger than the ultrasonic relaxation time and the nmr correlation time; in one group of liquids it is only slightly larger (2.5-5:1), but in another group it is considerably larger (100:1). There is an interesting observation with respect to distribution of times: for the case in which the dielectric relaxation time is about equal to the other times, a distribution of dielectric relaxation times is required, but where the dielectric time is much larger than the others a single time suffices.

Richard E. Nettleton of the Bureau of Standards began his presentation "The Phenomenology of Liquid Transport" by stressing the traditional aim of irreversible thermodynamics: that of providing a unified way of regarding constitutive relations. Take the problem of writing the most general stress-strain relation for a viscoelastic material, or that of determining relations among the elastic constants. The Onsager reciprocity relation may be applied here. Why, then, have not the relations among phenomenological coefficients which may be obtained from Onsager's theorem (and the Gibbs entropy equation) found much application to the numerical evaluation of these coefficients? The reason appears to be that there are other more direct means available; as an example, for chemical reactions, the kinetic coefficients are all readily calculable from the model.

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EDGERTON, GERMESHAUSEN & GRIER, INC. BOSTON • LAS VEGAS • SANTA BARBARA a model, and here the Onsager reciprocity relation may be useful. Take the irreversible approach to the steady states of heat conduction and diffusion. Over times of about 10^{-13} sec, Fourier's and Fuchs' laws may be augmented by inertial terms proportional to the time derivatives of the heat and particle flows. In this case, the generalized law of heat conduction can be obtained from a Debye-wave model of thermal propagation, but the terms associated with thermal diffusion can best be obtained from Onsager reciprocity, which obviates the need for doubtful assumptions about molecular motion in a fluid. Specifically, theoretical expressions may be obtained for the thermal conductivity and the thermal diffusion coefficient.

Shirley V. King of Birkbeck College, London, one of the coworkers of J. D. Bernal of the University of London in the geometrical approach to liquid structure, then presented a film on an aspect of close-packed spheres. Bernal's group has done some exceedingly interesting work on the statistics of close-packed spheres, and the film showed a case in which a shallow pan was filled with several layers of ball bearings and then agitated in a random manner. For whatever reason (and it is a matter of some controversy) structure began to emerge in the assemblage in the form of an area having definite crystalline form. This area then enlarged as the random agitation continued. Miss King did not have time to discuss this phenomenon sufficiently to give any kind of a complete explanation. But it is to be noted that it is not necessary to conclude that it illustrates order emerging almost miraculously from disorder by random agitation. One of the findings of Bernal in his studies of close-packed spheres is the distinction between "heaps" and "piles", the heap representing a disorderly arrangement, and the pile an orderly arrangement, of many objects thrown together. In the experimental case in question, the random agitation may be said to have enabled a "heap" to become a pile, it being presumed that the more regular packing is encouraged by agitation. The author is admittedly on dangerous ground here, but this very rough explanation has been attempted, since close-packed sphere theory is a particularly fundamental, as well as fascinating, field of physics.

Ernest Yeager of Western Reserve University began the chemistry session with a very fundamental discussion of the various effects associated with the propagation of ultrasonic waves through electrolytic solutions. In recent years, the relaxational absorption observed for many electrolytes has been explained quantitatively in terms of specific chemical processes including ionization, hydrolysis, ionic association, and even rearrangement of solvent molecules bound to ionic associates. Often several processes are perturbed simultaneously from equilibrium by the sound waves, and the interpretation of the resulting complex relaxation spectra requires considerable insight into the nature and coupling of the processes. Dr. Yeager reviewed the normal reaction coordinate approach to the description of the relaxation spectra of coupled processes. Some ions of low charge density depress rather than increase the ultrasonic absorption. An explanation for this depression was proposed on the basis of the Hall two-state model for the structure of water. The principal effect responsible for the depression of the absorption is believed to be a decrease in the energies of activation for the interconversion of the two (or more) structures.

Gordon Atkinson of the University of Maryland next spoke on ultrasonic absorption in electrolytes. He stated that the present theories of electrolytic solutions, based on the Debye model of rigid spheres in a continuum solvent, are inadequate, and that one is forced to consider specific solvent effects. One of the promising techniques for the examination of such effects is ultrasonic absorption. He gave a brief description of application of ultrasonic findings to interpretation of the dynamics of electrolytic systems, in particular varieties of relaxation mechanisms which were useful in interpretation. He examined ultrasonic absorption results in $MnSO_4$ solutions in detail, and found a consistent interpretation in terms of a three-step association process in a manner first proposed by Eigen.

Frank T. Gucker, of the University of Indiana, traced the relationship of various thermodynamic properties of solutions. Among those included in the discussion were the molar enthalpy, molar heat capacity, volume, compressibility, and free energy. The changes of these quantities with concentration and other parameters of the solution are important to an understanding of the fundamental theory of solutions. If a very accurate density measurement and a very accurate measurement of the velocity of sound can be made, the compressibility can be computed to the same accuracy, and valuable information concerning the other parameters can be gained, particularly if temperature and pressure variation is also employed. Gucker described two velocity-determining systems of very great precision. As an example of the precision, fifteen measurements of the velocity of sound in water at a temperature of 35°C showed a standard deviation of 0.002 percent.