Properties of Liquid Helium 3

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THE DISCOVERY of He^3, the rare stable isotope of helium, by Alvarez and Cornog in 1939, was of considerable interest to low temperature physicists, because it presented a possible test of the theory that many of the remarkable properties of liquid He^4 are due to the existence of an even number of fundamental particles in the He^4 atom. According to this theory the behavior of liquid He^3, with an odd number of fundamental particles in the atom, would be expected to be quite different from that of liquid He^4. The present article summarizes the low-temperature research on He^3 and its significance in the liquid helium problem.

Liquid He^4. Before the methods of obtaining He^3 and the experimental work performed with it are described, a brief account will be given of some of the properties of liquid He^4 and of the theory that stimulated the study of He^3. More detailed accounts of the properties and theories of liquid He^4 may be found elsewhere (*1–3*).

Between its critical point, 5.2° K, and 2.19° K liquid He^4 exhibits no unusual properties, with the exception that its viscosity decreases slightly as the temperature decreases. At 2.19° K (the "lambda-point") a transition to a new liquid phase with many unique properties occurs. This phase is called He II to distinguish it from He I, the liquid phase that is stable above the lambda-point. The transition takes place without a latent heat or a volume change and hence is a second-order transition, in contrast to the usual first-order phase transitions such as vaporization or solidification. The term lambda-point was suggested by the similarity of the shape of the heat capacity curve near this temperature to the Greek letter λ.

The properties of He II have caused it to be termed a superfluid. Its viscosity as determined by the measurement of the flow through fine capillaries becomes vanishingly small as the temperature is lowered. Furthermore, the heat conductivity becomes abnormally large—under suitable conditions a thousand times that of copper at room temperature. Perhaps an even more remarkable property is the propagation of heat pulses through He II by a wave motion, which is called "second sound," by analogy with the transmission of pressure differences in ordinary sound. Still another characteristic of He II is the mobile film that apparently defies gravity. For example, the film flows up over the rim of an empty beaker partially immersed in a He II bath until the liquid levels inside and outside the beaker are equalized. Finally, in contrast to all other substances, He^4 remains liquid when cooled under its own vapor pressure to absolute zero, and at that temperature it requires a pressure of 25 atmospheres for solidification.

It has been possible to correlate many of these phenomena by assuming that He II is composed of two interpenetrating fluids, a normal and a superfluid (*4*). The former, as the name implies, is assumed to be similar to an ordinary liquid, whereas the latter is assumed to have zero viscosity, zero entropy, and zero heat content. In this theory the relative amount of the normal and superfluid components is determined by the temperature, the fraction that is superfluid increasing rapidly from zero at the lambda-temperature to unity at 0° K. The flow through narrow channels is explained as being due primarily to the superfluid, which flows with zero viscosity, whereas the normal fluid is held back by viscous drag. The decrease of the viscosity as the temperature is lowered is due to the decrease in the proportion of the normal fluid. The mechanism of heat transfer is depicted as an internal convection of normal and superfluid, the former moving toward the cold zone and the latter toward the hot zone. This process, which occurs with much greater efficiency than ordinary heat conduction, results in the extremely high heat conductivity. Undoubtedly a major triumph of the two-fluid model was the prediction of second sound several years before its observation.

The two-fluid hypothesis, as first developed by Tisza (*4*), was inspired by a suggestion of F. London (*5*) that there is a connection between the lambda-transition in liquid He^4 and a peculiar condensation that can occur in a gas following Bose-Einstein statistics. In such a gas, starting at a characteristic temperature, T_0, the atoms begin to occupy the lowest energy state, and at 0° K all the atoms are in this state. At T_0 a maximum appears in the heat capacity of a Bose-Einstein gas; similarly there is a maximum in the heat capacity of liquid He^4 at the lambda-temperature. When the density of liquid He^4 is used in this gas model, T_0 is calculated to be 3.14° K, in reasonable agreement with the observed lambda-temperature, 2.19° K. It should be pointed out, however, that quantitative agreement is not to be expected, because the interactions between the atoms have been neglected in treating the liquid as a gas. In the development of the two-fluid theory, Tisza identified the superfluid with the atoms in the lowest energy state.

It is important to note that this condensation cannot occur in a system that follows Fermi-Dirac statistics, because the latter permits only a small number of atoms to occupy the lowest energy state. The statistics followed by a system of weakly interacting identical atoms is simply determined by the number of fundamental particles in each atom. If the number is even, as in He^4, Bose-Einstein statistics is obeyed; if it is odd, as in He^3, Fermi-Dirac statistics is obeyed. No further information regarding these types of statistics is needed to appreciate their significance in the helium problem. Clearly, any theory that bases the behavior of liquid He^4 on Bose-Einstein statistics cannot apply to He^3. The comparison, then, of liquid He^3 with liquid He^4 offers a means of evaluating the importance of statistics in influencing macroscopic behavior.

Sources of He^3. The natural abundance of He^3 is very small, varying from about 1 ppm in atmospheric helium to about 0.1 ppm in gas well helium. Although its separation from natural sources seems to be a formidable task, significant quantities of helium containing as much as 4 per cent He^3 have been obtained. The principal technique that has been used for the separation is thermal diffusion, and enrichment methods that take advantage of the superfluid properties of He^3–He^4 solutions (to be discussed later) have also been employed.

Fortunately, it became possible to avoid this difficult isotope separation by making use of the following nuclear reactions:

$$Li^6 + n \rightarrow H^3 + He^4 \qquad (1)$$

$$H^3 \xrightarrow{12.5\text{ yr}} He^3 + e^- \qquad (2)$$

Tritium (H^3) is first produced by irradiating lithium with neutrons in a chain-reacting pile, as shown in Eq. (1). The tritium is easily separated from the He^4—for example, by diffusion of the tritium through a heated palladium thimble—and is then set aside to decay (Eq. [2]). Finally, the He^3 that has grown in is separated from the remaining tritium. The isotopic purity of the He^3 is limited only by the completeness of the initial separation of tritium from He^4. The preparation of He^3 by this method was first reported by A. Novick (*6*) in his determination of the half-life of tritium.

Liquefaction of He^3. The question of immediate interest, once pure He^3 was obtained, was whether it would liquefy. This question was raised because the attractive forces in He^4 are so weak that they are barely able to overcome the zero-point energy to form a liquid phase. In He^3 the attractive forces are the same, but the zero-point energy is larger as a result of the smaller mass, and therefore it seemed possible that He^3 might not liquefy. Notwithstanding, de Boer and Lunbeck (*7*) predicted the vapor pressure and the critical point of liquid He^3 on the basis of a quantum theory of corresponding states. Their predictions were in excellent agreement with the first measurements on liquid He^3 by Sydoriak, Grilly, and Hammel (*8*) and with the subsequent measurements by the authors (*9*).

TABLE 1

VAPOR PRESSURES OF LIQUID He^3 AND LIQUID He^4

T (°K)	P_{He^3} (mm Hg)	P_{He^4} (mm Hg)	P_{He^3}/P_{He^4}
1.0	8.7	0.12	72
1.5	50.7	3.59	14
2.0	152	24.2	6.3
2.5	334	78.5	4.3
3.0	618	183	3.4
3.35	890	293	3.0

A comparison of the vapor pressures of the two liquids at several temperatures is given in Table 1; the boiling points, critical constants, and molar volumes (*10*) are given in Table 2. It is interesting to note that the greater zero-point energy of He^3 causes the molal volume of liquid He^3 to be much greater than that of liquid He^4.

TABLE 2

PHYSICAL CONSTANTS OF LIQUID He^3 AND LIQUID He^4

	He^3	He^4
Boiling point (°K)	3.20	4.22
Critical temperature (°K)	3.35	5.20
Critical pressure (mm Hg)	890	1718
Molal volume at 1°K (ml)	38	27.6
Lambda-temperature (°K)	?	2.19

Flow properties of liquid He^3. After it had been established that He^3 would liquefy, the authors (*11*) made a search for superfluidity by comparing the flow rate of liquid He^3 with that of liquid He^4 through a narrow channel, or "superleak." The leak was constructed by shrinking a Pyrex glass capillary around a platinum wire of 0.005 in. diameter. On cooling, a narrow annular channel was formed because of the difference between the coefficients of expansion of the glass and the metal. By measuring the flow rate of

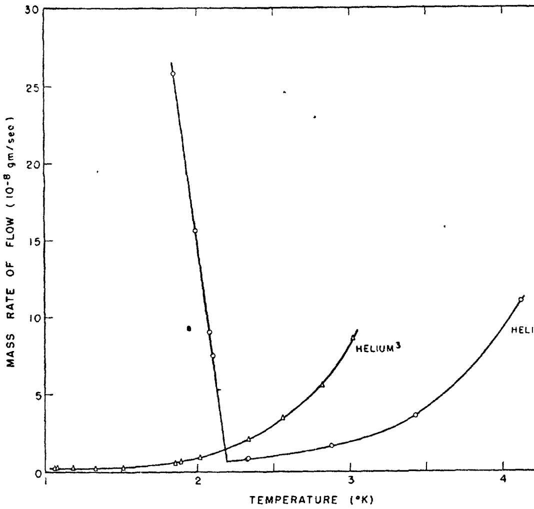

FIG. 1. Mass rate of flow of liquid He^3 and of liquid He^4 through a superleak, as a function of temperature.

helium gas through the leak at 4.22° K, the width of the channel was estimated to be 7×10^{-5} cm. The assembly was in the shape of a long U-tube supported vertically in a liquid helium bath, with the superleak constituting the lower 5 cm of one leg. The experiments were performed by condensing about 0.02 ml of liquid at the top of the leak and then permitting the liquid to flow through under its own vapor pressure. The amount of material flowing through the leak was observed as a pressure rise in a large bulb outside the helium bath. As shown in Fig. 1, the mass rate of flow of liquid He^4 was observed to decrease with temperature until the lambda-point was reached, and then to increase sharply with further decrease in temperature. On the other hand, the rate of flow of liquid He^3 under the same conditions decreased steadily as the temperature was lowered from 3.2° K to the lowest temperature attainable in the apparatus—1.05° K.

It was thus established that superfluidity does not occur in liquid He^3 down to 1.05° K. This temperature is probably low enough for the conclusion to be drawn that liquid He^3 does not become superfluid at any temperature, in agreement with the theory that the lambda-transition in He^4 is connected with Bose-Einstein statistics. The conclusion was further corroborated by an experiment of Daunt and Heer (*12*) with He^3–He^4 solutions, which will be described later. From their results they deduced that pure He^3 is not superfluid down to at least 0.25° K.

The viscosity of liquid He^3 has been obtained from measurements of the flow rate through the superleak under various pressure heads (*13*). It was found that the viscosity increases from a value of 22 micropoise at 2.8° K to 30 micropoise at 1.0° K. By comparison, the viscosity of He I decreases slightly from 33 micropoise at 5° K to 30 micropoise at 2.7° K and then more rapidly to 22 micropoise at the lambda-point, just as though the He^4 were anticipating the lambda-point. The fact that the viscosity of liquid He^3 increases with decreasing temperature is therefore believed to be further evidence for the nonexistence of a superfluid transition in He^3.

Solidification of He^3. The absence of superfluidity in He^3 raised the question whether He^3 would remain liquid at absolute zero. An examination of the melting pressure curve of He^4 seems to indicate that there would have been a triple point at 1° K if the lambda-point had not occurred. Instead, the melting pressure curve suddenly changes course at the lambda-point and then rapidly approaches a constant value of 25 atmospheres. It seemed possible, therefore, that liquid He^3, which has no lambda-point, might solidify under its own vapor pressure.

A schematic diagram of the apparatus used by the authors (*14, 15*) to determine the melting curve of He^3 is given in Fig. 2. The technique employed was the blocked capillary method used by Keesom to determine the melting pressure of He^4. As the pressure in the system was slowly raised by compressing the He^3 gas in the reservoir (*H*) with the hydraulic system (*I*), the two Bourdon gauges (*B, G*) read the same until solidification occurred. Then, because of the plug of solid that now blocked the capillary U-tube (*D*), gauge (*B*) remained constant at the melting pressure, whereas gauge (*G*) continued to increase. Conversely, as the pressure was lowered, the gauges became equal again at the melting pressure. For measurements between 1.5° K and 1.02° K the capillary U-tube was immersed directly in a bath of liquid helium (*E*) boiling under reduced pressure, and for measurements down to 0.16° K it was surrounded by a paramagnetic salt (ferric ammonium alum), which was cooled by adiabatic demagnetization.

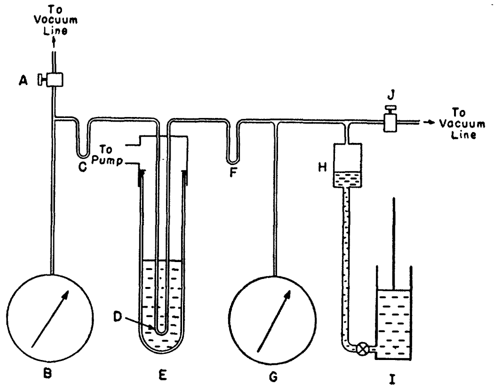

FIG. 2. Apparatus for the measurement of the melting pressure of He^3 (schematic).

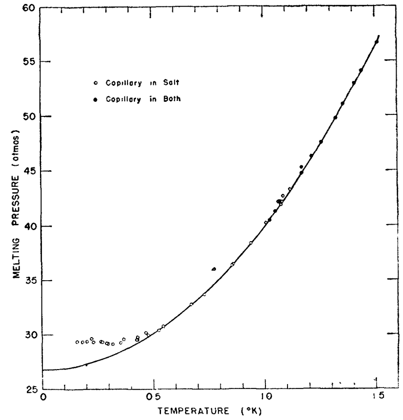

FIG. 3. Melting pressure of He^3 vs. temperature.

The results of both sets of measurements are plotted in Fig. 3. From 0.5° K to 1.5° K the data are represented by the equation

$$P = 26.8 + 13.1\ T^2 \text{ atm}, \qquad (3)$$

which is shown as the solid curve in the figure. Below 0.5° K the observed pressures approach a constant value of 29.3 atmospheres, but there is a possibility that the deviation of these points from Eq. (3) may be due to poor thermal contact between the capillary and the salt below 0.5° K. It is clear, however, that the melting pressure is at least 26.8 atmospheres at absolute zero, and that He^3, like He^4, remains liquid under its own vapor pressure at absolute zero.

Nuclear alignment in He^3. Besides yielding information about the physical state of He^3 near absolute zero, the melting pressure curve can answer the question of nuclear alignment in the liquid. At high temperatures the nuclear spins in He^3 (equal to ½) are randomly oriented, and this disorder contributes to the entropy of both liquid and solid. At sufficiently low temperatures the spins may become aligned in a parallel or antiparallel arrangement, with a concomitant loss in the nuclear spin entropy. This effect would be absent in He^4, since it has no nuclear spin. Pomeranchuk (*16*) has predicted that, because of exchange forces effective in the liquid but not in the solid, this alignment will take place in the liquid near 1° K but not in the solid until $10^{-7\circ}$ K. According to his prediction the entropies of the liquid and solid become equal near 1° K, and below this temperature (down to $10^{-7\circ}$ K) the entropy of the liquid is less than that of the solid. Consequently, the melting pressure curve would have a minimum at 1° K and then rise as the temperature is lowered further. As was shown in Fig. 3, however, the experimentally determined melting pressures do not show a minimum, in disagreement with Pomeranchuk's prediction, and it can be concluded that nuclear alignment does not occur in the liquid down to 0.5° K. Further experimental work is needed to determine the shape of the melting pressure curve below 0.5° K, for it is still possible that nuclear alignment will occur at a lower, experimentally realizable temperature.

He^3–He^4 solutions. A discussion of He^3 is not complete without consideration of some of the properties of He^3–He^4 solutions. It will be possible to cover only the highlights of the extensive research with these solutions, but a more comprehensive treatment may be found in a recent review article (*17*).

It was suggested by Franck (*18*) that the He^3 in a He^3–He^4 solution would not participate in superflow and that an enrichment of He^3 could be obtained by making use of this property. The idea was first tested by Daunt, Probst, Johnston, Aldrich, and Nier (*19*) in a film flow experiment with atmospheric helium. A Dewar vessel, capped with a ground-glass plug, was partially immersed in liquid He II, so that the surface of the bath was below the cap. When energy was supplied to an electrical heater in the Dewar vessel, helium flowed in through the ground-glass joint solely by means of the mobile film. The helium that collected in the Dewar was shown by mass spectrometer analysis to have a He^3 concentration of less than 5×10^{-8}, as compared to the starting concentration of 1.2×10^{-6}. The result indicated that, in agreement with Franck's suggestion, He^3 does not partake of the superflow in the helium film and that it is indeed possible to enrich He^3 by taking advantage of this behavior.

Further evidence that the He^3 in liquid helium does not participate in superflow was obtained by Lane, Fairbank, Aldrich, and Nier (*20*). In their apparatus, shown schematically in Fig. 4, a heater (*H*) was placed in the interior of an inverted bulb (*B*), which communicated with the filling system through a glass capillary U-tube (*C*), a Kovar tube (*K*), and another glass capillary (*T*). The whole assembly was immersed in a bath of liquid helium contained in a Dewar (*D*), and then atmospheric helium was condensed in the bulb and U-tube to the level indicated. With the bath below the lambda-point, heat was supplied to the heater, and 10 minutes later a portion of the liquid was removed from the Kovar tube by evaporation. The sample thus obtained contained 50 per cent of the total quantity of He^3 in the system, and the concentration of He^3 in the sample was 130 times the original value. This observation is explained in terms of the two-fluid model of heat conduction as a "heat flush," in which the superfluid atoms move toward the heater, whereas the He^3 atoms move away from the heater in company with the normal fluid and thus concentrate in the Kovar tube.

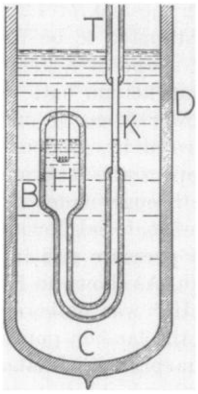

FIG. 4. Apparatus for the enrichment of He^3 in He^3–He^4 solutions by heat flush (schematic).

The heat flush technique has been incorporated in several of the procedures for the concentration of He^3 from natural sources. For example, Reynolds, Fairbank, Lane, McInteer, and Nier (*21*), starting with helium enriched to 0.01 per cent He^3 by thermal diffusion, have increased the concentration to 4 per cent. Using the heat flush method, Eselson and Lazarev (*22*) have produced helium containing 0.01 per cent He^3 from helium with an initial He^3/He^4 ratio of 5×10^{-8}, and Taconis has produced helium with .5 per cent He^3 from atmospheric helium with a He^3 content of 1.2×10^{-6}.

The postulate that He^3 associates only with the normal component of He^4 was extended further by Taconis, Beenakker, Nier, and Aldrich (*23*) in the interpretation of their vapor pressure measurements with a He^3–He^4 mixture containing 0.05 per cent He^3. The vapor pressure of an ideal solution is described by Raoult's law,

$$P = xP_3^\circ + (1 - x)P_4^\circ, \tag{4}$$

where P is the vapor pressure of the solution, P_3° and P_4° are the vapor pressures of pure He^3 and pure He^4, respectively, and x is the mole fraction of He^3 in the solution. Offhand, one would expect that a mixture of two isotopes would form an ideal solution. However, the observed values of P below the lambda-temperature were always higher than predicted by Raoult's law, and the deviation increased as the temperature was lowered. A satisfactory explanation was obtained by assuming that the He^3 should be considered as dissolved only in the normal fluid and that x should be calculated without counting the superfluid component. As the temperature is lowered below the lambda-point, the amount of the normal fluid decreases, and hence x, the mole fraction of He^3 in the normal fluid, increases. This concept has been embodied in de Boer and Gorter's theoretical treatment (*24*) of the properties of He^3–He^4 solutions, and subsequent measurements have indicated that it is at least qualitatively correct.

In all the experiments with He^3–He^4 solutions described thus far, the He^3 content has been too small to cause a measurable change of the lambda-temperature of He^4. The lowering of the lambda-temperature by the addition of He^3 was first demonstrated by the authors (*25*) in another experiment with the superleak previously described. In these measurements the criterion for the occurrence of the lambda-transition in a solution was a break in the plot of mass rate of flow vs. temperature, similar to that shown for pure He^4 in Fig. 1. The results obtained in this way are shown as squares in Fig. 5. Daunt and Heer (*12*) extended the measurements to higher concentrations, using the appearance of the abnormal heat conductivity through the He II film to locate the lambda-temperatures. Their results, shown as open circles in Fig. 5, gave 0.38° K as the lambda-temperature of a solution with 89 per cent He^3, and, by extrapolation of the measured curve to 100 per cent He^3, they concluded that pure He^3 cannot be superfluid above 0.25° K. Also shown in Fig. 5 is a point for 1.5 per cent He^3 determined by Eselson, Lazarev, and Lifshits (*26*), who observed the temperature at which flow through the helium film begins. As a consequence of the decrease in lambda-temperature with increasing He^3 concentration, it is not possible to make pure He^3 by methods based solely on superfluidity. At temperatures that can be conveniently reached by pumping on liquid helium (about 1° K) the limit is approximately 60 per cent He^3.

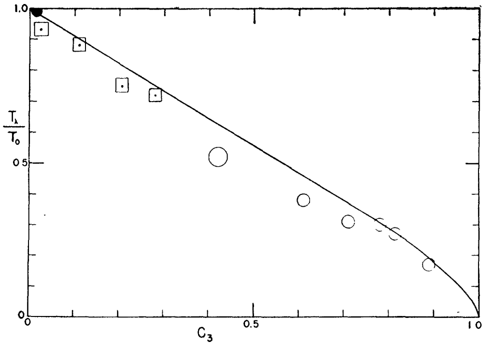

FIG. 5. Ratio of the lambda-temperatures of He^3–He^4 solutions, T_λ, to the lambda-temperature of pure He^4, T_0, as a function of the mole fraction of He^3, C_3. ● Eselson, Lazarev, and Lifshits (*26*) ; □ Abraham, Weinstock, and Osborne (*25*) ; ○ Daunt and Heer (*12*).

The apparent association of He^3 with only the normal fluid suggested that a He^3–He^4 solution below its lambda temperature might separate into two physically distinct liquid phases. The authors (*27*) have proved, however, that only a single liquid phase exists below the lambda-point by showing that the vapor pressures of a 20.3 and of a 25.5 per cent solution of He^3 in He^4 are different at all temperatures down to 1.04° K, well below the lambda-temperatures of both solutions. The proof follows from the phase rule, which would require the two solutions to have the same vapor pressure at a given temperature, if each had separated into two liquid phases. The existence of a single liquid phase above and below the lambda-point has the consequence that the lambda-transition in solutions is second order, as it is in pure He^4.

In conclusion it must be pointed out that, although the absence of superfluidity in He^3 shows that the lambda-transition in liquid He^4 is connected with the Bose-Einstein statistics, the theoretical explanation of the properties of liquid helium is far from complete. No mention has been made of Landau's (*28*) theory of liquid helium, which does not depend on statistics, but which, in some respects, is more successful than Tisza's (*4*) theory in explaining the properties of He II. Since Landau's theory makes no distinction between He^3 and He^4, it seems desirable to remove the last vestige of doubt about a superfluid transition in He^3 by investigating the flow properties of the *pure* liquid below 1° K. It is surprising that no experimental evidence has yet been found for nuclear alignment in liquid He^3; additional information on this point can be obtained by extending the melting curve below 0.5° K and by measuring the specific heat and heat of vaporization of the liquid.

From the experimental comparison of the two isotopic liquids, He^3 and He^4, it would almost seem as though they were different elements, and it must be concluded that the statistical behavior of the atoms—Fermi-Dirac for He^3 and Bose-Einstein for He^4—does indeed affect the macroscopic behavior.

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News and Notes

Scientists in the News

Lowell Besley, professor of forestry and dean of the Forestry School at the University of British Columbia, has been named executive director–forester of the American Forestry Association, effective July 1.

Robert W. Cairns, assistant director of research for the Hercules Powder Company, has been named vice-chairman of the Department of Defense Research and Development Board. He has been associated with the board since 1949 as a member of a panel of the Committee on Ordnance.

Hans T. Clarke, professor of biochemistry at the College of Physicians and Surgeons, Columbia University, gave the first A. J. Goldforb annual lecture, entitled "Fifty Years of Progress in Experimental Biology." The series of public lectures was endowed by former students and associates in honor of Dr. Goldforb, professor emeritus of biology, City College, New York.

Robert L. Cushing has been named director of the Pineapple Research Institute of Hawaii. He has been assistant director since 1951, and acting director since June 1952.

A scroll of appreciation was presented to **William Dameshek** for his teaching in hematology and his inspiring leadership over the past 25 years, by former and present associates on the occasion of the Advanced Course in Hematology at New England Center Hospital, Boston.

William von E. Doering, Yale professor of chemistry, who helped to synthesize quinine, will receive the $1000 American Chemical Society Award in Pure Chemistry in Los Angeles on Mar. 16.

Hugo C. Duzan, William R. McCallum, and **Thomas R. Todd** have received the Highway Research Board Award, given annually for an outstanding technical paper. The selection was made for their paper "Recent Trends in Highway Bond Financing," presented at last year's annual meeting of the board, and published in the *Proceedings*.

Ben Fisher has resigned as assistant director of hematologic research, Michael Reese Hospital, Chicago, to accept the newly created position of clinical pathologist at Wayne County General Hospital, Eloise, Mich.

George Gibson has been named acting chairman of the Chemistry Department at Illinois Institute of Technology during a nine-month leave of absence granted to **Martin Kilpatrick**, chairman.

Rolf Gran-Olsson, of the Norwegian Institute of Technology in Trondheim, will be visiting professor at Brown University in the Graduate Division of Applied Mathematics for the spring semester. He will teach courses in advanced elasticity and methods of technical mechanics.

Grayson L. Kirk, vice president and provost of Columbia University, has been named to succeed **Dwight D. Eisenhower** as president of the university. General Eisenhower became president-emeritus and trustee-emeritus, effective Jan. 19.

George A. Kolstad has been appointed chief of the Physics and Mathematics Branch in the AEC's Division of Research. Dr. Kolstad has been a physicist with the AEC in Washington since 1950.

Cheng Chun Lee has joined Eli Lilly and Company as a pharmacologist in the Toxicology Department. Dr. Lee, a native of Youngchow, Kiangsoo, China, received his Ph.D. from Michigan State College last year.

William S. Lohr, head of the Department of Civil Engineering, **Paul B. Eaton**, head of the Department of Mechanical Engineering, and **Luther F. Witmer**, of the Department of Metallurgical Engineering, will retire from the faculty of Lafayette College in June, each having served more than 30 years.

Charles H. Mann has been appointed medical director of Bristol Laboratories Inc. For the past three years he has served as medical director for the Heyden Chemical Corp.

Georg Masing, of the Institut für Allgemeine Metallkunde, Universitat Göttingen, has received the Institute of Metals (Platinum) Medal for 1953, for