and S<sup>32</sup>. We note that the average binding energy per particle for the three shells is, respectively, 8.3, 9.3, and 8.6 Mev.

In connection with the computed mass values of Barkas,<sup>2</sup> there are several misprints and errors which have apparently been propagated widely throughout the literature. The values for O<sup>19</sup> and O<sup>20</sup> seem to be in error, along with the entire group of A = 4n+2, T = 2 nuclei, for which the wrong symmetry character was used in making the calculations. In Table I, corrected values are reported for these nuclei and compared with probable experimental values taken from the table of isotopes prepared by Seaborg and Perlman.<sup>6</sup> The agreement is improved in every case. A more comprehensive study of the agreement of the Wigner theory with new experimental data of  $\beta$ -decay energies and Q-values will be presented by the author in the future.

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## Angular Distribution of Annihilation Radiation

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WHILE testing with annihilation radiation a circuit selecting the coincident pulses from two scintillation counters, it was realized that the angular correlation between the two annihilation photons could easily be measured with far greater accuracy than previously reported.<sup>1</sup> As a consequence, we performed several measurements on the angular distribution of the two photons, using sources of Cu<sup>64</sup> surrounded by a gold absorber in which the positrons were stopped.

During the final measurements, whose results are reported in Fig. 1, the distance between the source and either detector was 120 cm. The source and detectors were aligned on the same horizontal line by means of a cathetometer, which was also used to measure the vertical displacements of one of the detectors. The detectors, seen from the source, subtended a vertical angle of  $4 \times 10^{-3}$  rad, while the source, seen from the detectors, covered an angle about ten times smaller.

The zero on the abscissas of Fig. 1. corresponds to the position where the source and the upper surfaces of the detectors were in the same horizontal line. The coincidence rate has a maximum for a



FIG. 1. Gamma-gamma-coincidences as a function of the position of one of the detectors.

negative abscissa, corresponding to the position where the line joining the centers of the detectors passes through the source. The fact that the counting rate does not vanish for positive displacements definitely indicates that the annihilation gamma-rays are not always emitted in exactly opposite directions.

If one approximates with an exponential the tail of the curve for positive abscissas, one obtains for the average value of the momentum of the center of mass of the annihilating pairs approximately  $0.9 \times 10^{-2}$  in units of mc. This is remarkably close to the value of  $0.8 \times 10^{-2}$  obtained by Dumond, Lind, and Watson,<sup>2</sup> from the shape of the annihilation line from a Cu absorber.

A more detailed description of the experiment and a more complete theoretical discussion is under preparation.

\* Assisted by the joint program of the ONR and the AEC.
<sup>1</sup> R. Beringer and C. G. Montgomery, Phys. Rev. 61, 222 (1942).
<sup>2</sup> Dumond, Lind, and Watson, Phys. Rev. 75, 1226 (1949).

## Erratum: The Second Viscosity of Liquids [Phys. Rev. 75, 1415 (1949)]

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HE values for n' given in column 5 of Table I are incorrect and should be deleted. It was intended that this column be derived by multiplication of columns 3 and 4 and the correct values are easily obtainable in this way.

## Anomalous Adsorption of Helium at Liquid Helium Temperatures

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URING measurements on the distribution of He<sup>3</sup> between gas and adsorbed film for dilute mixtures of He3 and He4 adsorbed on jeweler's rouge (Fe<sub>2</sub>O<sub>3</sub>) in the He II region,<sup>1</sup> we encountered equilibrium pressures markedly different from those previously reported.2 We, therefore, made preliminary measurements of the adsorption isotherms for He4.

The adsorbent was 0.81 g of Fe<sub>2</sub>O<sub>3</sub>, of surface area 1.7 m<sup>2</sup>, as determined from a Kr isotherm at 90.2°K.

The data are shown in Fig. 1, in which the volume adsorbed in S.T.P. cc is plotted against the ratio of the measured equilibrium pressure P to the saturation pressure  $P_0$  of the bulk liquid He at the measuring temperature. The dotted curve is the isotherm at 2.45°K; the solid curve is for all measured temperatures below  $T_{\lambda}$ , namely, 2.11°, 1.78°, and 1.53°K.<sup>3</sup>

The data present several unusual features:

(1) The He II isotherms are identical, within our error of 2 percent between 1.53° and 2.11°K, and from 25 to 80 percent saturation. Consequently, the heat of vaporization of the adsorbed He in this region must equal that of the bulk liquid (see also Frederikse, reference 3). As the adsorbed He is in thermal equilibrium with vapor at  $P < P_0$ , this result requires that the entropy of the adsorbed layers be higher than that of liquid He, as predicted several years ago by Kramers.<sup>4</sup> The heat of adsorption would have to be changed by about twice our experimental error in order to account for the apparent entropy difference.

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(2) Application of the simple BET theory<sup>5</sup> shows that the volume adsorbed in the first layer  $(v_m)$  is much larger than that calculated from surface areas and the liquid density, as already shown by Schaeffer et al.<sup>6</sup> from their data for carbon black at 4.2°K. We find for  $v_m$  about 0.9 cc/m<sup>2</sup>, four times the value calculated from the liquid density, and corresponding to an atomic spacing in solid He under several hundred atmospheres pressure. The data of Schweers<sup>7</sup> at small  $P/P_0$  also fit a BET plot, giving about the same  $v_m$  on glass and O<sub>2</sub> below 2.5°K, and yield also the value of Schaeffer et al.  $(v_m = 0.57 \text{ cc/m}^2)$  at the b.p. It seems, therefore, certain that the first adsorbed layer on most surfaces is highly compressed.

Schweers' stated result, that the adsorption energy drops by ca. 50 cal./mole in building up the first four layers, must now be re-interpreted as representing the work of compression necessary to complete the mono-laver.8

(3) The He II isotherms show anomalously high adsorption, starting at  $P/P_0 \sim 0.7$ , and increasing enormously at high saturations. We reached the limiting value of 40  $cc/m^2$  with a distinctly lower equilibrium pressure than  $P_0$ . This value yields a minimum film thickness of  $\sim \! 160$  atomic layers, on the reasonable assumption of liquid-type spacing (4A) for all but the first few layers.9

We consider this anomalous adsorption as due to the formation of the Rollin film. Previously widely differing results<sup>10</sup> on the film thickness are explainable because of the extreme sensitivity of the film thickness to percent saturation.

We observed some indication of slight hysteresis in desorption above  $P/P_0=0.9$  below  $T_{\lambda}$ , whereas in all other parts of the isotherm the equilibrium pressures were identical in adsorption and desorption.

The isotherm at 2.45°K yields a  $v_m = 0.9 \text{ cc/m}^2$ , the same as below  $T_{\lambda}$ , but shows distinctly smaller adsorption above  $P/P_0$  $\sim$ 0.20, reaching saturation with  $\sim$ 30 layers adsorbed.

The two methods proposed by Harkins and Jura<sup>11</sup> for plotting adsorption data  $(\log P/P_0 vs. 1/v^2)$ , and the "film pressure"  $\pi vs.$ the area  $\sigma$  available per atom adsorbed) show a distinct break at  $P/P_0 \sim 0.7$ , in a manner which in their treatment would indicate a two-dimensional second-order transition. This, along with the



disappearance of the effects above  $T_{\lambda}$ , suggests that the phenomenon is cooperative, and of the same character as the  $\lambda$ -point in bulk liquid He.

We plan to investigate in more detail the anomalous density of the first adsorbed layers, the "transition" at  $P/P_0 \sim 0.7$  with the resulting high adsorption, and the entropy of the system as a function of the number of adsorbed lavers.

function of the number of adsorbed layers. <sup>1</sup> Measurements made in collaboration with Professor A. O. C. Nier and Mr. B. B. McInteer, Department of Physics, University of Minnesota. <sup>2</sup> J. Kistemaker, Leiden Comm. No. 271-b. <sup>3</sup> The He 11 results are in good agreement with unpublished data of H. P. R. Frederikse, Leiden, who kindly sent us his data on the isotherms between 1.39° and 1.99°K, up to 85 percent saturation. We are indebted to him for these data, and for the manuscript of his communication to Physica on the specific heat of adsorbed He on FeoD<sub>3</sub>, prior to publication. <sup>4</sup> H. A. Kramers, personal communication to L. M. <sup>5</sup> Brunauer, Emmett, and Teller, J. Am. Chem. Soc. **60**, 309 (1938). <sup>6</sup> Schaeffer, Smith, and Wendell, J. Am. Chem. Soc. **71**, 863 (1949). <sup>7</sup> J. Schweers, Thesis, Leiden, 1941. <sup>8</sup> Suggested by W. Band. <sup>9</sup> Kistemaker's statement that the number of adsorbed layers does not increase over ~30 layers, above  $P/P_0 \sim 0.9$ , is due to his method of evalua-tion, only valid below  $P/P_0 \sim 0.8$ , as it assumes the number of adsorbed layers to be equal in the two adsorption bulbs he compares. His original data show the effect we observed. <sup>10</sup> See W. H. Keesom, *Helium*, Elsevier Publishing Company, Inc., New York, 1942) pp. 292-308. <sup>11</sup> See Geo. Jura and W. D. Harkins, J. Am. Chem. Soc. **68**, 1941 (1946).

## Anomalous Adsorption of Helium at Liquid **Helium Temperatures**

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HE results reported recently<sup>1</sup> under the above title by Earl A. Long and Lothar Meyer raise the following question: The "Brunauer-Emmett-Teller" theory2 assumes that the saturation number of atoms in each monolayer is a constant and should be roughly equal to the number to be expected from the interatomic spacing in the liquid phase. However, by applying the B.E.T. isotherm to their observed adsorption for helium, Long and Meyer find that in the neighborhood of the lambda-temperature the saturation number in the first monolayer is about four times that expected from the liquid density. Schaeffer et al.<sup>3</sup> similarly found about twice the expected number at the boiling point temperature. Can these results be considered real, or are they simply an indication that the B.E.T. theory breaks down completely?

The density of liquid helium is unusually low at these temperatures because of the zero point energy, and it is, therefore, physically reasonable to suppose that the potential energy of adsorption can compress the first, and perhaps partly also the second monolayer to be adsorbed. Moreover the measurements by Frederickse4 on the specific heat of an amount of adsorbed helium that corresponds to the first (compressed) monolayer, are actually consistent with that of a two-dimensional Debye solid with a characteristic temperature of about 18°K. It is, therefore, reasonable to consider the film at low pressures as immobilized, and to apply the following generalization of the B.E.T. theory: Let the number of sites available to the second monolayer be only some fraction  $\alpha_1$  of the number of atoms adsorbed in the first monolayer, the number of sites available to the third monolayer be another fraction  $\alpha_2$  of the number of atoms adsorbed in the second layer, and so on. This problem can be solved by the methods introduced by T. L. Hill.<sup>5</sup> In the simplest case where only one anomalous monolayer is present, i.e.,  $\alpha_1 < 1$ , while  $\alpha_2$  etc. are all = 1, the resulting adsorption isotherm is a simple generalization of the B.E.T. equation.

$$(V_1/V)\{x/(1-x)\} = \{x+\beta_1(1-x)^{\alpha_1}\}/\{1-x(1-\alpha_1)\},$$
 (1)