

Assuming a value² of 0.32080 for the ratio $\nu(B^{11})/\nu(H^1)$, one obtains

$$\nu(Nb^{93})/\nu(H^1) = 0.24441 \pm 0.00013.$$

This value should be increased by a Lamb³ correction of 0.38 percent because of the diamagnetic effects of the atomic electrons. Use of this correction and of the value 5.58504 for the g -factor of the proton⁴ leads to the following value for the nuclear g -factor of Nb^{93} :

$$g(Nb^{93}) = 1.3702 \pm 0.0007.$$

Since the spin of Nb^{93} is known from hyperfine structure data⁵ to be $9/2$, the nuclear magnetic moment of Nb^{93} is

$$\mu(Nb^{93}) = 6.1659 \pm 0.0032 \text{ nuclear magnetons.}$$

These values are to be compared with the earlier values $g=1.18$ and $\mu=5.31$ obtained by Meeks and Fisher⁶ from hyperfine structure studies. The value obtained in the present work lies close to the upper Schmidt limit value of $+6.79$ for a $g_{9/2}$ odd proton, which Nb^{93} has according to Mayer's⁷ theory.

We wish to express our appreciation to the Ohio State University Development Fund and Research Foundation for grants that made this work possible.

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¹ J. R. Zimmerman and D. Williams, *Phys. Rev.* **76**, 350 (1949).

² W. H. Chambers and D. Williams, *Phys. Rev.* **76**, 638 (1949).

³ W. E. Lamb, *Phys. Rev.* **60**, 817 (1941).

⁴ J. E. Mack, *Rev. Mod. Phys.* **22**, 64 (1950).

⁵ S. S. Ballard, *Phys. Rev.* **46**, 806 (1934).

⁶ W. W. Meeks and R. A. Fisher, *Phys. Rev.* **72**, 451 (1947).

⁷ M. Mayer, *Phys. Rev.* **75**, 1969 (1949).

Isotope Effect in the Superconductivity of Mercury*

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THE existence of a small quantity of Hg^{198} at the National Bureau of Standards¹ prompted us to investigate its properties as a superconductor.² The sample available to us had a high degree of isotopic separation and was approximately 98 percent pure Hg^{198} . The average atomic weight³ of natural mercury is 200.6. The mercury had been produced by the transmutation of gold and had been prepared by distilling it off the bombarded gold foil.

Preliminary results are now available on the critical field behavior and transition temperature. The destruction of superconductivity was detected magnetically by a ballistic galvanometer method and the zero-field transition temperature determined by extrapolation of the critical field curve to zero field. (Further details of the experimental method will appear elsewhere.) Tests were made with natural mercury and with two specimens of Hg^{198} about 400 mg each (both derived from the original sample). The natural mercury was prepared by our Chemistry Division and is presumed to contain less than 0.001 percent impurity. The specimens of Hg^{198} were separately redistilled in vacuum (following the original distillation from the gold foil) and were enclosed in Pyrex capillaries which were evacuated and sealed off. Temperatures were measured with a helium vapor pressure thermometer using the tables prepared at the Royal Society Mond Laboratory (June 4, 1949) for reducing the pressures to degrees Kelvin.

The results are indicated in Fig. 1 which is a plot of current in the Helmholtz coils (at critical field) vs. the absolute temperature for both the natural mercury and Hg^{198} . Any small difference in slope is not significant since the demagnetization factors of the samples and the exact disposition in the field may have been different. The intercepts are significant and give the transition temperatures as 4.156°K for natural mercury and 4.177°K for Hg^{198} .

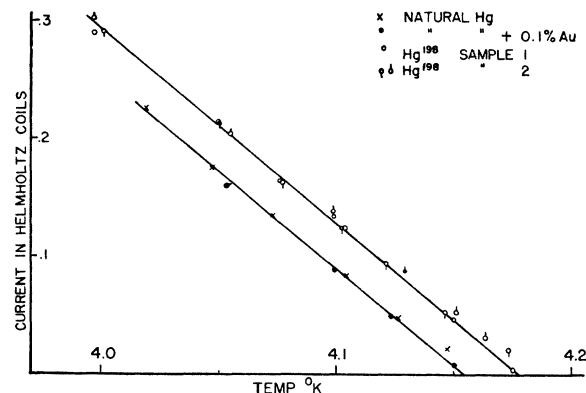


FIG. 1. Current in the Helmholtz coils at the critical field vs. the absolute temperature.

Many pains were taken to exclude the possibility of secondary effects. The earth's field was compensated to within 10^{-4} gauss. The distillation apparatus and capillaries were carefully cleaned. The clean appearance of the mercury was good evidence of the absence of base metal impurities in excess of 0.001 percent but there was some possibility that the Hg^{198} might have contained a gold impurity, derived from the original foil which would not have shown up in the form of surface sum. Spectrochemical analysis of the Hg^{198} was not feasible, so a sample of natural mercury with 0.1 percent gold was prepared and tested. (A saturated solution of gold in mercury will contain about 0.15 percent.) The results are given by the black dots and show that even this large amount of gold impurity is unimportant.

From these results one may infer that the transition temperature of a superconductor is a function of the nuclear mass, the lighter the mass the higher the transition temperature. At the ONR conference in Atlanta, March 20-21, it was reported that Serin and Reynolds⁴ at Rutgers University had undertaken a similar investigation independently and simultaneously with our own. They found that the transition temperatures of Hg^{199} , Hg^{202} , and Hg^{204} were also a function of the atomic mass, with the lighter isotopes having higher transition temperatures. It is gratifying to find that our results are mutually consistent and complementary. Taken together they definitely establish a dependence of transition temperature on mass number.

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¹ We are indebted to Dr. W. F. Meggers for making this sample available to us.

² The suggestion of examining a pure isotope was also made to us independently by Professor K. F. Herzfeld.

³ A. O. Nier, *Phys. Rev.* **52**, 933 (1937).

⁴ Reynolds, Serin, Wright, and Nesbitt, *Phys. Rev.* **78**, 487 (1950).

Magic Numbers and Elements with No Stable Isotopes

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THAT the abnormal instability of *all* isotopes of $_{43}Tc$ and $_{81}Pm$ may have something to do with the occurrence, close by, of magic neutron numbers 50 and 82, is an obvious enough suggestion. It becomes even more plausible if we consider that only the absence of odd-mass isotopes requires a special explanation (even-mass isotopes are, for $Z=43$ and 61, excluded by parity rules). Two other elements, A and Ce, then appear to share the same anomaly and in both the latter cases the connection with the magic neutron numbers (20 and 82, respectively) is even more evident.^{1,2}

These cases of abnormal instability can also be seen as an anomaly in the pattern of stability of the adjacent nuclei. The stable pairs $\text{Cl}^{37}/\text{K}^{39}$, $\text{Mo}^{97}/\text{Ru}^{99}$, $\text{La}^{139}/\text{Pr}^{141}$, and $\text{Nd}^{145}/\text{Sm}^{147}$ represent the transition from an odd-valued A to $A+2$ by adding *two protons* (a “ pp step” for short). The occurrence of such steps, for nuclei of odd mass, however, is incompatible with the simple assumptions underlying the semi-empirical formulas of Weizsäcker and his followers,³ according to which there exists for each A an idealized value of Z (or of $I=N-Z$) which “should” correspond to maximum stability so that the isotopic number I_{nat} of any naturally stable nucleus of an odd mass A “should” be the odd integer nearest to the *idealized* value $I_{\text{id}}=f(A)$. If this picture is true, then on an $A-I$ diagram all odd- A nuclei lie on successive horizontal segments which begin and end at those abscissae for which I_{id} is an even integer. For example, between A corresponding to $I_{\text{id}}=4$ and that corresponding to $I_{\text{id}}=6$, all stable odd-mass nuclei must have $I_{\text{nat}}=5$. Since the $I_{\text{id}}=f(A)$ function is steadily rising, two successive odd-mass nuclei can differ either by one nucleon of each kind (an “ np step,” or a step along the horizontal), or by two neutrons (an “ nn step,” or a step leading from the end of a horizontal to the beginning of the next one). There must be no backsliding into a lower valued horizontal; that is, no “ pp steps.”

Figure 1 shows all stable odd-mass nuclei from $A=71$ to 171, together with the $I_{\text{id}}=f(A)$ curve according to reference 3 (one of its original constants, however, has been adjusted in order to improve the fit). The scale of I is exaggerated in order to emphasize the transitions between horizontals. We see that at the *beginning* of each horizontal there is a surfeit of neutrons; it is there that any perturbing factors, favoring the addition of protons, are most likely to produce a pp step instead of a normal np one.

In the case of the magic number $N=20$ the nature of the disturbance is clear. The first odd-mass nucleus which contains exactly 20 neutrons, that is Cl^{37} , also happens to be the first on the horizontal $I=3$. Since the 21st neutron is supposed to be bound very weakly, the competition of a pp step, always strong at the beginning of a horizontal, becomes overwhelming and we find the next odd-mass nucleus, K^{39} , back on the horizontal 1, in contradiction to the normal Weizsäcker picture.¹

This step brings back the condition of neutron deficiency, as it exists at the end of a horizontal; the next step should be nn , or possibly an np if the binding of the 22nd neutron as well is abnormally weak (since both competing steps contain the 21st neutron, the weakness of the latter's binding is irrelevant). No such continued abnormality seems to exist, since with K^{41} the horizontal 3 is resumed, and hereafter no further deviations are observed until the completion of the next neutron shell. We may conclude that, after the closing of the shell $N=20$, only one neutron is bound weakly enough to cause visible perturbation.

In the case of the shell $N=82$, the completion takes place at La^{139} . Since this nucleus is the *second* on its horizontal ($I=25$), and not the first, as in the previous case, the competition of a pp step is less pronounced. The binding of the 83rd neutron is, however, weak enough to make an np step unable to stand even this competition. Accordingly, we find the next odd-mass nucleus, Pr^{141} , on the horizontal 23. One more step is taken *along* this lower horizontal, leading to Nd^{143} , which shows that the abnormal weakness of binding extends to at least one more open-shell neutron.

The completion of the shell $N=50$ looks, however, quite different. Here the odd-mass nucleus concerned (Y^{89}) happens to be the *fifth* on its horizontal ($I=11$), in a region of neutron deficiency (below the I_{id} line) rather than surfeit as in the previous cases. The chances of a pp step toward a still lower horizontal appear to be nil, but the weakness of the binding of the open-shell neutrons (51st, 52nd, etc.) favor np steps against nn . Accordingly, the horizontal 11 is abnormally long and leads into a region of an abnormally pronounced neutron deficiency. An nn step leading to Mo^{97} ($I=13$) relieves this deficiency almost completely, but before the horizontal 13 asserts itself, we have, with Ru^{99} , a last

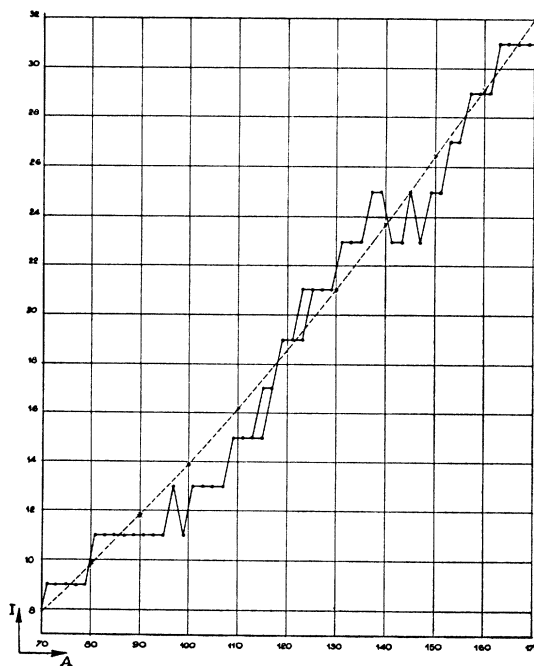


FIG. 1. Dotted line: $I_{\text{id}} = A(A^3 - 0.7)/(A^3 + 128)$.

reappearance of the horizontal 11. To explain this abnormal step (which is responsible for the instability of Tc), we have no longer the resource of postulating a very weakly bound neutron (56th, which would be analogous to the 21st in the argon case) sandwiched between two more strongly bound ones: the 55th neutron does *not* close a shell. And yet it seems tempting to look for some property of open-shell neutrons (this description should cover half a dozen, or so, neutrons attached after the completion of the shell $N=50$) which would be responsible for this anomaly.

Short of devising waves or kinks in the sequence of binding energies for open-shell neutrons, we suggest one *ad hoc* hypothesis. The usual parity rules make no distinction of principle between odd- Z and odd- N nuclei; if we start, for example, from an odd- N nucleus, then an np step, which pairs off the neutrons and introduces an odd proton, is no more and no less likely, from the point of view of these rules, than an nn or a pp step which do not change parity at all. We suggest that in the region of open-shell neutrons, this equivalence does not hold: for these loosely bound neutrons, the pairing-off does *not* bring the full normal benefit in binding energy and therefore *cannot* be balanced against the pairing-off of protons. Starting from an odd- N nucleus, the competition of *both* nn and pp steps is strengthened; and as the competition is strongest, respectively, at the end and at the beginning of a horizontal, we must expect it to become operative at the transition between horizontals. In other words, if a horizontal begins in a region of open-shell neutrons, we may expect to find there a tendency toward the elimination of stable isotopes of odd Z . By the time the end of the next horizontal is reached, the next neutron shell is built far enough to allow the resumption of normal parity rules.

The natural instability of Tc and Pm thus gives us a glimpse of the conditions governing the binding of open-shell neutrons (comparable in a way to the open-shell electrons of strongly metallic elements). Other glimpses can be derived from:

(1) The asymmetric fission of uranium. The rule already mentioned by M. G. Mayer⁴ can be conveniently formulated as follows: “the most probable modes of fission are those which lead to a minimum number of open-shell neutrons,” this number being obtained by adding such neutrons from *both* fragments.

(2) The general trend of capture cross sections for slow neutrons. There seems to be little doubt that these cross sections are, in the aggregate, noticeably lower in the regions of open-shell neutrons (say, $A=88$ to 100 and 138 to 148) than in the regions starting with Rh and Sm. One would say that the addition of neutrons is "inhibited" in the weak-binding region, and then "stimulated" by the accumulated lag behind the rising trend of the idealized Weizsäcker curve. Such crude considerations may help in finding the way to reconcile the droplet and the shell concepts.

I am indebted to Dr. B. T. Feld and Professor J. A. Wheeler for valuable discussions.

- ¹ H. Bethe and R. Bacher, *Rev. Mod. Phys.* **8**, 177 (1936).
² N. Popelka and W. D. Harkins, *Phys. Rev.* **77**, 756 (1950).
³ H. Bethe and R. Bacher, reference 1, p. 166.
⁴ M. G. Mayer, *Phys. Rev.* **74**, 235 (1948).

General Operator Field Equations Derived from A Variation Principle. Construction of "Divergence-Less" Four-Vector Operator

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IT was shown in a recent report¹ that commutator equations for fields, in particular Maxwell's field equations in vacuum, could be derived from a variation principle. The Lagrangian was constructed by replacing the classical space-time derivatives of the field variables by i/\hbar multiplied into the respective commutators of the displacement operators and the field variables. The operator field equations resulted when the first variation of the trace of the Lagrangian with respect to the matrix elements of the field variables was made to vanish. It is of interest to generalize this procedure to general invariant functions constructed from four displacement operators p_μ and any number of field operators f_A , $A=1, 2, \dots$, without putting emphasis upon the replacement of partial derivatives by corresponding commutators.

The arbitrary invariant function of the p_μ and f_A will be denoted by $L(p_\mu, f_A)$ and its trace by G , with

$$G = \text{Tr}[L(p_\mu, f_A)]. \quad (1)$$

Now the complete first variation of G is

$$\delta G = [\partial G / \partial (p_\mu)_{mn}] \delta (p_\mu)_{nm} + [\partial G / \partial (f_A)_{mn}] \delta (f_A)_{nm} \quad (2)$$

$$= (N^\mu)_{mn} \delta (p_\mu)_{nm} + (F^A)_{mn} \delta (f_A)_{nm}, \quad (3)$$

where N^μ and F^A are accordingly defined. It is well known, however, that the trace of a matrix, say M , is invariant under transformations of type

$$M' = e^{iS} M e^{-iS}. \quad (4)$$

Consequently, if one subjects L in (1) to a transformation of type (4) and takes S to be an arbitrary infinitesimal operator of the first order, then we have to the desired order

$$\delta (p_\mu) = i[S, p_\mu], \quad \delta (f_A) = i[S, f_A], \quad (5)$$

so that,

$$\begin{aligned} \delta G &= i(N^\mu)_{mn} [S, p_\mu]_{nm} + i(F^A)_{mn} [S, f_A]_{nm} \\ &= i[P_\mu, N^\mu]_{mn} S_{nm} + i[F_A, f^A]_{mn} S_{nm}. \end{aligned} \quad (6)$$

But the field equations are

$$F^A = 0, \quad (7)$$

and the transformation (4) was such that $\delta G = 0$. One concludes then that

$$[p_\mu, N^\mu] = 0. \quad (8)$$

As a summary, we have shown that a field theory based on the traces of an arbitrary invariant function of the four displacement operators p_μ and any number of field operators f_A , $A=1, 2, \dots$, has associated with it the four-vector operator N^μ , defined in (3), which satisfies the "continuity" equation, (8).

As an example, let us consider the field equations for a scalar field U based on the Lagrangians L_+ or L_- defined by

$$L_\pm = \frac{1}{2} [p_\mu, U]_\pm [p^\mu, U]_\pm,$$

where $[p_\mu, U]_+$ denotes the anti-commutator expression and $[p_\mu, U]_-$ the usual commutator expression:

$$[p_\mu, U]_\pm \equiv [p_\mu, U \pm U p_\mu].$$

From (2) and (3), the field equations are

$$[p_\mu, [p^\mu, U]_\pm]_\pm = 0,$$

and the 4-vector operators are

$$N_\pm^\mu = [U, [U, p^\mu]_\pm]_\pm.$$

It can be shown that $[p_\mu, N_\pm^\mu] = 0$. Preliminary investigations indicate that N^μ has the character of a 4-current "particle" density operator. Similar expressions can be calculated for fields based upon Lagrangians of types resembling those in current use. There are no classical analogs, however, for fields based on Lagrangians which contain anti-commutator expressions or, more generally, for those fields based on Lagrangians which have the p_μ entering in an arbitrary way.

- ¹ C. Gregory, *Phys. Rev.* **78**, 67 (1950).

Masses of Pd¹⁰⁴, Pd¹⁰⁸, Pt¹⁹⁵, and Cu⁶⁵ *

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A DOUBLE-FOCUSING mass spectrograph has been used to photograph a number of doublets formed by singly charged hydrocarbon molecules and multiply charged palladium and platinum atoms. These photographs have been used to obtain precise values for the masses of Pd¹⁰⁴, Pd¹⁰⁸, and Pt¹⁹⁵.

Masses of Pd¹⁰⁴ and Pd¹⁰⁸.—A high frequency spark between two palladium electrodes served as the source of palladium ions. The quadruply charged Pd¹⁰⁴ and Pd¹⁰⁸ atoms formed wide doublets at mass numbers 26 and 27 with singly charged C₂H₂ and C₂H₃. The hydrocarbons were secured in the manner described previously.¹ From fourteen photographs of the C₂H₂—Pd¹⁰⁴ doublet, the difference in packing fraction between C₂H₂ and Pd¹⁰⁴ was found to be $\Delta f = 15.34 \pm 0.04$, with extreme values of 15.05 and 15.60. From seventeen photographs of the C₂H₃—Pd¹⁰⁸ doublet, the packing fraction difference was found to be $\Delta f = 17.74 \pm 0.03$, with extreme values of 17.48 and 17.97. The spacing between the two hydrocarbons was used as the mass scale in these measurements. Using K. T. Bainbridge's recommended mass values² for H¹ and C¹², the packing fractions of C₂H₂ and C₂H₃ are found to be $f = 9.218 \pm 0.017$ and $f = 11.887 \pm 0.018$, respectively. These values, combined with the above packing fraction differences, give for Pd¹⁰⁴, $f = -6.12 \pm 0.05$, and for Pd¹⁰⁸, $f = -5.85 \pm 0.04$.

It was planned to check these values by photographing the Pd¹⁰⁴—Cr⁵² and Pd¹⁰⁸—Fe⁵⁴ doublets at mass numbers 52 and 54, and combining the results with the Cr⁵² and Fe⁵⁴ masses reported previously.¹ From nine photographs, the Pd¹⁰⁴—Cr⁵² packing fraction difference was found to be $\Delta f = 2.31 \pm 0.04$, with extreme values of 2.08 and 2.46. These doublets were very unsymmetrical, the Cr⁵² line being very much the heavier. From twelve photographs of the well-matched Pd¹⁰⁸—Fe⁵⁴ doublet, $\Delta f = 2.25 \pm 0.02$, with extreme values of 2.16 and 2.37. On eight of these photographs, well-matched Pd¹⁰⁶—Cr⁵³ and Pd¹¹⁰—Mn⁵⁵