Mass Dependence of the Superconducting Transition Temperature of Mercury*

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IN a recent communication,[1] Herzfeld, Maxwell, and Scott fitted the available data on the superconducting transition temperatures (T_c) of mercury isotopes of different mass (M), to a formula of the type $M^{\alpha}T_c=$const. by the method of least squares. The best fit to the[2] data was given by $\alpha=0.378$, whereas in an earlier communication,[3] we had proposed $\alpha=\frac{1}{2}$.

At the time of our communication, we did not feel that the accuracy of the data warranted a close scrutiny of the kind given to it by Herzfeld and co-authors; and the exponent $\frac{1}{2}$ was proposed as giving a good fit within the accuracy of the experimental values for the transition temperatures. The poor quality of the data is evident from the appreciable deviations of the experimental points from the relation, $M^{0.378}T_c=$const., derived by least squares.

Since our communication, the detailed theories of superconductivity of Bardeen[4] and Fröhlich[5] give the value $\frac{1}{2}$ for α.

Because of the importance of determining an accurate experimental value of α, in view of these theories, we have been engaged in an extensive investigation of the transition temperatures of mercury samples of different isotopic constitutions. We have found that the temperature spread of the transition from the superconducting to the normal state (and thus the accuracy with which the zero-field transition can be determined by extrapolation) is very sensitive to the way in which the samples are cooled to liquid air temperature, and also to the way in which liquid helium is admitted to the flask. The procedure finally adopted was to place a small amount of liquid air in the outer shield flask. The system was allowed to stand for about an hour, and then small amounts of liquid air were added over the course of another hour. It was possible to get an accurate idea of the temperature of the samples during cooling by measuring the change in resistance of pick-up coils surrounding them; and thus to bring the samples slowly through the freezing point of mercury. We found it equally important never to permit the initial blow-off of warm helium gas from the transfer tube to hit the samples; thus, we waited until liquid helium poured out of the transfer tube before placing the tube in the helium flask. This method of cooling clearly produces the minimum amount of strain in the samples.

Our most reliable data for our original samples and for a new set of three samples[6] of slightly different isotopic constitution (making a total of seven samples) are shown in Fig. 1, where $\log_{10}M$ is plotted *vs.* $\log_{10}T_c$. The measuring technique was the same as that described earlier,[7] except that the frequency of the alternating field was reduced to 20 c.p.s. The transition temperature for the sample of natural mercury is in good agreement with the value, 4.167°K, given by Misener.[8]

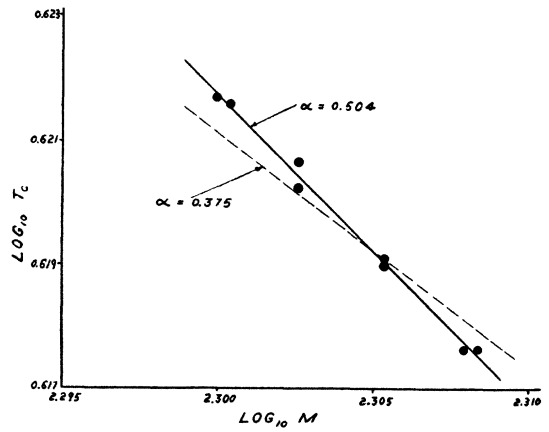

FIG. 1. Transition temperature as a function of the mass number.

The slope of the line as drawn in Fig. 1 is 0.504. For purposes of comparison a line of slope 0.375 is also drawn.

On the basis of these measurements, we feel that it is definitely established that in mercury, at least, the exponent α is certainly much closer to $\frac{1}{2}$ than to any other value having a simple physical interpretation. This result is in agreement with the predictions of the theories[4,5] mentioned above.

* This work was supported by the ONR, by the Research Corporation, by the Rutgers University Research Council, and by the Radio Corporation of America.
[1] Herzfeld, Maxwell, and Scott, Phys. Rev. **79**, 911 (1950).
[2] Herzfeld *et al.* conclude that the true value is $\alpha=3/8$, corresponding to constant thermal energy of the lattice.
[3] Serin, Reynolds, and Nesbitt, Phys. Rev. **78**, 813 (1950).
[4] J. Bardeen, Phys. Rev. **79**, 167 (1950).
[5] H. Fröhlich, Proc. Phys. Soc. London **63**, 778 (1950); Phys. Rev. **79**, 845 (1950).
[6] The isotopes were produced by Carbide and Carbon Chemical Division, Oak Ridge National Laboratory, Y-12 Area, Oak Ridge, Tennessee, and were obtained on allocation.
[7] Reynolds, Serin, Wright, and Nesbitt, Phys. Rev. **78**, 487 (1950).
[8] A. D. Misener, Proc. Roy. Soc. **174**, A, 262 (1940).

Temperature Effect in Geiger-Müller Counters

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THE temperature effect on the rate of spurious discharges in G-M counters (hard glass sealed-off type with out-gassed cathode of 2 cm diam. and 5 cm long) was observed when they were heated and cooled at constant rates. As is shown in Fig. 1, on the heating stage the rate of natural counts increased in various manners which depended on the filling gases and less markedly on the cathode material; it decreased rapidly on the cooling stage without exception. It was verified that these discharges were due to "Spontanentladung" and the occurrence of double or triple discharges ("Nachentladung") was very rare. The plateau curves were taken with a gamma-ray source and also without any source before and after each experiment, and we could not find marked changes in them, though those with natural counts were generally better after heating. The plateau lengths were generally between 150 and 500 v, with the slopes of 0.6 to 10 percent per 100 v. We observed the increases in spurious counts also when the counter voltage was applied intermittently, that is, for 20 sec. once every 3 min. The increased counting rate began to decrease again when the temperature rise was stopped and the counter was kept at a certain constant temperature near 100°C. The decrease was an almost exponential function of the time measured from the

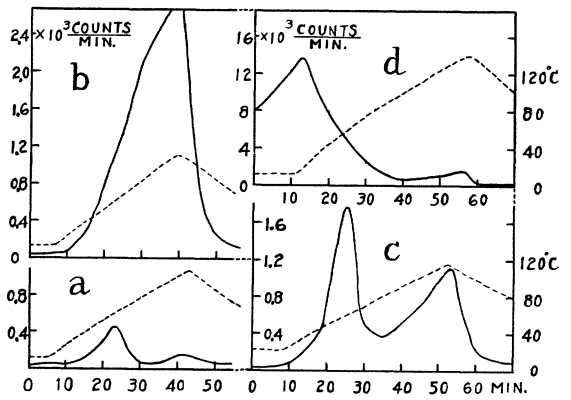

FIG. 1. Temperature effects on the rate of spontaneous discharges of the G-M counters. Full line: counts per min. The broken line gives the temperature. (a): A 76 mm, ethyl ether 9 mm. (b): A 95 mm, alcohol 10 mm. (c): A 98 mm, ethylene 10 mm. (d): A 90 mm, alcohol 10 mm, under visible light irradiation. All with Fe cathode and 0.1-mm W wire.

moment when the temperature rise was stopped. The above phenomena could not be attributed to the decrease of the work function of the cathode surfaces by heating, because (1) the counting rate was not a function of the temperature itself but of its sense and rate of change, and (2) the photo-sensitivity (by yellowish-red light from *W* filament lamp) decreased by heating as shown in Fig. 1a, though it increased gradually with time when the counter was kept at room temperature, as was found by Nonaka.[1] A tentative explanation can be given along the lines of adsorption and evaporation of the quenching gas molecules and the negative ions from the cathode surfaces.[2] A full account will appear in the *Journal of the Physical Society of Japan*.

[1] I. Nonaka, J. Phys. Soc. Japan **3**, 322 (1948).
[2] The author is indebted to Professor I. Nonaka for his kind discussion on these points.

The Gamma-Rays from Neutron-Activated Gold

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WITH the advent of the study of photo-electron and pair production "lines" in the scintillation gamma-ray spectrometer,[1] there arise many fruitful applications of this new technique. Some new results in the low energy region, where the photoelectric effect predominates, have been obtained with neutron-activated gold and I^{131}. In the present spectrometer arrangement use is made of a small 1-cm cube crystal of NaI-Tl exposed to the uncollimated gamma-rays from the source, and an E.M.I. 5311 photo-multiplier operating at 1000 volts.

Figure 1 shows the differential pulse-height distribution curves obtained with a neutron-activated gold sample of 0.1 mC a few days after the irradiation. In *A* the features are the 69-kev Hg x-ray below 10 volts, the well-known 411-kev Au^{198} gamma-ray below 40 volts, and the unresolved hump between 10 and 20 volts. This is believed (see below) to correspond to the 159-kev and 209-kev lines known to belong to 3.3-day Au^{199}, which is formed by two successive (n,γ) reactions on Au^{197} during the irradiation,[2] and

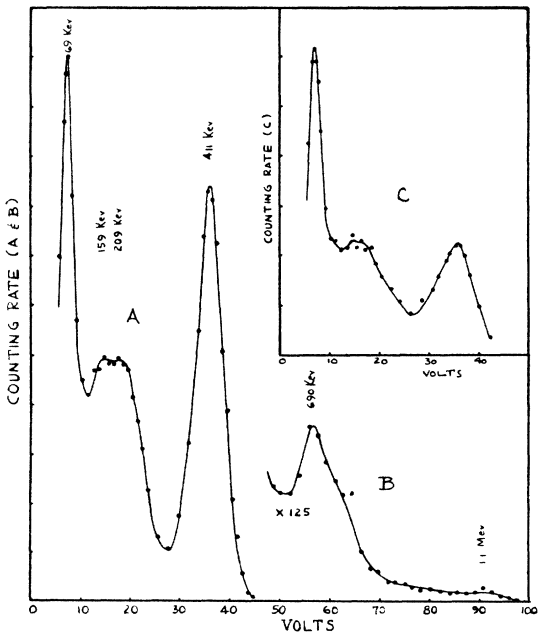

FIG. 1. Scintillation spectrometer pulse-height distributions for the gamma-rays from neutron-activated gold. *A*. Lower energy gamma-rays and x-ray. *B*. Counting rate scale ×125 to show new high energy components. *C*. Relative distribution of *A* after 11 half-lives of Au^{198}.

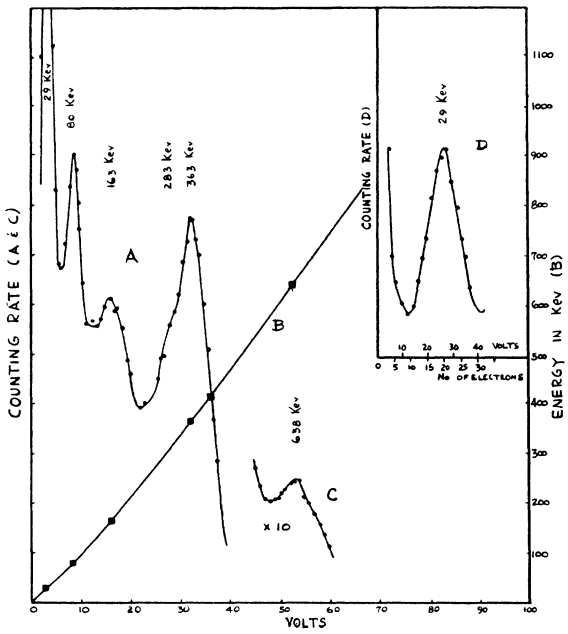

FIG. 2. Gamma-ray from I^{131}. *A*. Lower energy gamma-rays and x-ray. *B*. Energy-pulse-height relationship showing slight lack of linearity. *C*. Counting rate scale ×10 to show 638-kev component. *D*. X-ray at 29 kev shown on higher gain setting, with scale to show pulse heights estimated in terms of the number of electrons collected from the photo-multiplier cathode.

also to a contribution due to the Compton effect of the 411-kev gamma-ray. The lines occur at the full gamma-ray energies because of the trapping in the crystal of the low energy radiations and particles following the initial photoelectric effects. In *B* the counting rate scale has been magnified 125 times, and one can see clearly the presence of a new 690-kev gamma-ray below 60 volts and a new 1.1-Mev gamma-ray near 90 volts.[3] The calibration curve used to determine these energies is given in Fig. 2. In Fig. 1(*C*) the low energy region is given once again, this time after a lapse of 30 days from the observations of 1(*A*). The counting rate scale is given arbitrarily, but it is evident that the 2.7-day 411-kev component has decayed more rapidly than the rest of the curve. The decay rate for the curve below 20 volts agrees with that for a mixture of 3.3-day Au^{199} and 2.7-day Au^{198}. The relative sizes of the x-ray peaks in *A* and *C* suggest a large internal conversion coefficient for the Au^{199} 159-kev and 209-kev lines. Because of the much lower counting rates involved it has not been possible to follow the high energy components over more than 3 or 4 half-lives. The half-life of these appears, however, to be less than 3.0 days, and probably close to 2.7 days. One can therefore draw some interesting conclusions concerning the decay scheme for Au^{198}. The energy of the 411-kev line adds to the 690-kev line to give the component at 1.1 Mev, thus suggesting that the latter corresponds to a cross-over transition. We therefore visualize the existence of excited states in Hg^{198} at 411 kev and 1.1 Mev, and a complex beta-spectrum for Au^{198} which would consist of two components[4] of 960 kev and 270 kev (say two percent). This lower energy component has apparently not yet been detected.

The pulse-height-energy calibration curve for the scintillation spectrometer has been shown to be almost linear, but not exactly so. Figure 2 for I^{131} (gamma-rays 80 kev, 163 kev, 283 kev, 363 kev, and 638 kev, and x-ray 29 kev for Xe) illustrates the necessity of having such a curve (2(*B*)) for the instrument, using known gamma-ray energies. In the calibration curve a reference point corresponding to the Au^{198} 411-kev line is included. A slight curvature which is most marked below 150 kev may be observed, and thorough testing having shown that this curvature can only be