

These observations can be correlated on the basis of the ZnS:Cu crystal being an N -type semiconductor with donor levels distributed over an energy range and with an exhaustion barrier at each electrode. When a voltage is applied, the cathode barrier will widen as donor levels are emptied until most of the voltage appears across the cathode barrier. An ac applied potential V depletes and replenishes the two barrier layers alternately. Above a critical potential V_i quantum-mechanical tunneling through the cathode barrier occurs. Below V_i conduction electrons originate only within the crystal. With a local field larger than the field E_i necessary to ionize deep-lying donor levels and for $E_i > E_a$, the critical field for acceleration of conduction electrons, electrons can be extracted from these deep-lying levels and accelerated to kinetic energies great enough to impact excite activator ions. With $V < V_i$ only light out of phase with the voltage will appear; with $V > V_i$ electrons will tunnel with the electrodes producing additional light in phase with the voltage.

For example, assuming a semiconductor with a work function less than the metal electrode work function and with a homogeneous concentration of predominantly shallow donors, a Mott-Schottky barrier appears at each electrode. The local field E is linear with the distance from the barrier boundary reaching a maximum at the crystal surface. A two-volt barrier requires a field of 10^7 v/cm for appreciable penetration.⁵ For a donor concentration of 3×10^{17} cm⁻³, a dielectric constant of 10, and an applied potential of 10^3 volts, the required field for tunneling exists at the cathode surface. The barrier thickness is then 2×10^{-4} cm. Deep-lying donor levels 0.5 ev below the conduction band can be field excited with 10^6 v/cm, a field which exists at the surface when 10 volts is applied to the crystal and which is sufficient to accelerate conduction electrons.

Further experimental and theoretical work are in progress to test and refine the proposed mechanism.

† Presented in part March 22, 1952, at the Columbus Physical Society Meeting.

¹ William W. Piper, J. Chem. Phys. (to be published).

² G. Destriau, Phil. Mag. 38, 700 (1947).

³ Lehovec, Accardo, Jamgochian, Phys. Rev. 83, 603 (1951).

⁴ J. R. Haynes and H. B. Briggs, Phys. Rev. 86, 647 (1952).

⁵ Stern, Gossling, and Fowler, Proc. Roy. Soc. (London) 124A, 699 (1929).

The Effect of Atomic Binding on Nuclear Reaction Energies

R. SERBER

Columbia University, New York, New York

AND

H. S. SNYDER

Brookhaven National Laboratory, Upton, New York

(Received May 21, 1952)

THE energetics of nuclear reactions are commonly computed using atomic masses. In a reaction in which the nuclear charge changes, the difference in electronic binding energies of the final and initial atoms is thus properly taken into account (aside from the small energy difference between the final atom, which is charged, and the corresponding neutral atoms). There does, however, seem to exist some confusion¹ as to the mechanism of the transfer of electronic binding energy to the incoming or outgoing nuclear particles, despite the fact that this mechanism is of the simplest kind: the effect on a charged particle of the electrostatic field produced by the cloud of electrons.

Let us ask how the energy of an atom changes if the nuclear charge is changed from Z to $Z+dZ$. The reason there is a change is that the nucleus is in a region of negative electrostatic potential produced by the electrons; if we call the electrostatic potential produced by the electrons at the position of the nucleus $\varphi(Z)$, the energy change is just $edZ\varphi(Z)$, and the energy of the atom $E(Z)$ satisfies the equation

$$dE(Z)/dZ = e\varphi(Z). \quad (1)$$

Equation (1) is used in the Fermi-Thomas model of the atom to obtain $E(Z)$ from the calculated value of $\varphi(Z)$; conversely, (1) may be used to obtain $\varphi(Z)$ from an empirical determination of $E(Z)$. The Fermi-Thomas relation, with a constant adjusted to give agreement with the known energies of light atoms, is $E(Z) = -15.73Z^{7/3}$ ev, which gives

$$e\varphi(Z) = -36.70Z^{4/3} \text{ ev.} \quad (2a)$$

According to Foldy,² a better representation for heavy atoms is $E(Z) = -RZ^{12/5}$ ($R=13.6$ ev), which gives

$$e\varphi(Z) = -32.64Z^{7/5} \text{ ev.} \quad (2b)$$

If we consider, as an example, a case of β -decay, the β -particle at the nucleus is also in the potential $\varphi(Z)$, and gains from this a kinetic energy $-e\varphi(Z)$ in passing through the atom. This gain is, of course, just the negative of the change in atomic energy discussed above, and one sees quite clearly the mechanism by which the energy conservation law is satisfied.

The energy gain $-e\varphi(Z)$ does not represent a lower limit to the energy of the emerging β -particle. It must be remembered that the emerging β -particle feels the Coulomb field of the nucleus as well as the field due to the electrons, and its total potential energy as it passes through the atom is always negative. Thus there is no energetic reason why it should not come out with zero energy. The effect of the field due to the electrons on the shape of the β -spectrum is properly represented by using a screened Coulomb potential, rather than the potential of a bare nucleus, in calculating the Coulomb wave functions for the β -particle.³

The foregoing remarks are not quite accurate, in that the change in nuclear charge was considered infinitesimal, rather than a multiple of e , and any effect of a redistribution of the electron cloud between initial and final atom was thus ignored. If, in any nuclear reaction, the charged particles entering or leaving the nucleus have velocities high compared to the electronic orbital velocities, the electron cloud will not have time to readjust, and the particles feel just the electronic potential of the initial atom. The net energy transfer to incoming and emerging nuclear particles, due to their interaction with the atomic electrons, is then just $-e(Z'-Z)\varphi(Z)$, where Z' is the atomic number of the final nucleus. On the other hand, if the process were adiabatic, the energy transfer to the nuclear particles would be $-[E(Z')-E(Z)]$. The latter difference can be evaluated by integration of (1) with respect to Z ; it represents the energy change if the nuclear charge is altered by infinitesimal steps, allowing the electron cloud to readjust after each step. The difference

$$\Delta E = -[E(Z')-E(Z)] + e(Z'-Z)\varphi(Z) \quad (3)$$

would therefore be lost to the emerging nuclear particles in a completely nonadiabatic transition; this energy appears as excitation energy of the final atom. In any particular event the final atom is left in a definite stationary state, and the energy loss to the nuclear products is the energy difference between this state and the ground state. There will be a probability distribution for ending in any given final state, and ΔE represents the average energy loss, averaged over this probability distribution. In a partially adiabatic transition, the mean excitation energy of the final atom will of course be less than ΔE .

If $E(Z')-E(Z)$ is expanded in a Taylor series in $Z'-Z$, we obtain

$$\Delta E = -\frac{1}{2}(Z'-Z)^2 d^2 E(Z)/dZ^2 = -\frac{1}{2}e(Z'-Z)^2 d\varphi(Z)/dZ. \quad (4)$$

Using (2a) or (2b) we find

$$\Delta E = 24.47Z^{1/3}(Z'-Z)^2 \text{ ev,} \quad (4a)$$

$$\Delta E = 22.85Z^{2/5}(Z'-Z)^2 \text{ ev.} \quad (4b)$$

For $Z=90$, $|Z'-Z|=1$, (4b) gives $\Delta E=138$ ev, whereas $-e\varphi(90)=17.8$ kev.

Another simple estimate of ΔE can be made by using hydrogenic wave functions for each of the electrons. An electron in a state of principal quantum number n , which feels an effective nuclear charge Z_{eff} , contributes to $E(Z)$ an amount $-Z_{\text{eff}}^2 R/n^2$, to

$E(Z')$ an amount $-(Z_{\text{eff}}+Z'-Z)^2R/n^2$, and (using the virial theorem) to $e\varphi(Z)$ an amount $-2Z_{\text{eff}}R/n^2$. The total contribution to ΔE is just $(Z'-Z)^2R/n^2$. Thus each closed shell of principal quantum number n adds $2R(Z'-Z)^2=27.2(Z'-Z)^2$ ev to ΔE . For $|Z'-Z|=1$, $Z=10$, this gives $\Delta E=54$ ev, while (4a) and (4b) give 53 ev and 57 ev. For $Z=90$ the simple recipe gives $\Delta E=125$ ev, and (4a) and (4b) give 110 ev and 138 ev.

A formal treatment of the arguments given above is quite simple. Let P_n be the probability that the electronic system of the final atom will be left in a state of energy $E_{Z'n}$. If $n=0$ denotes the ground state, the mean excitation energy of the atom will be

$$\Delta E = \sum_n P_n (E_{Z'n} - E_{Z'0}).$$

For a completely nonadiabatic transition from initial to final atom,

$$P_n = (\psi_{Z'n}, \psi_{Z'0})|^2,$$

where the $\psi_{Z'n}$ are the wave functions of the states of the final atom, $\psi_{Z'0}$ is the wave function of the ground state of the initial atom, and

$$(\psi_{Z'n}, \psi_{Z'0}) = \int \psi_{Z'n}^* \psi_{Z'0} d\tau.$$

If the Schrödinger equation for the initial atom is

$$E_{Z'0}\psi_{Z'0} = H_Z\psi_{Z'0},$$

that for the final atom is

$$E_{Z'n}\psi_{Z'n} = \left[H_Z - \sum_{i=1}^Z \frac{(Z'-Z)e^2}{r_i} \right] \psi_{Z'n},$$

and we can write

$$\begin{aligned} \Delta E &= \sum_n (\psi_{Z'0}, \psi_{Z'n}) (\psi_{Z'n}, \psi_{Z'0}) (E_{Z'n} - E_{Z'0}) \\ &= \sum_n (\psi_{Z'0}, \psi_{Z'n}) \left(\psi_{Z'n}, \left[H_Z - \sum_{i=1}^Z \frac{(Z'-Z)e^2}{r_i} - E_{Z'0} \right] \psi_{Z'0} \right). \end{aligned}$$

The closure theorem can be used to evaluate the sum over n , and we obtain

$$\begin{aligned} \Delta E &= - \left(\psi_{Z'0}, \sum_{i=1}^Z \frac{(Z'-Z)e^2}{r_i} \psi_{Z'0} \right) - (E_{Z'0} - E_{Z'0}) \\ &= (Z'-Z)e\varphi(Z) - (E_{Z'0} - E_{Z'0}), \quad (5) \end{aligned}$$

in agreement with (4). The quantity ΔE is, by its definition, positive. Thus if $E_{Z'0} - E_{Z'0}$ is expanded in a Taylor series in powers of $Z'-Z$, the term linear in $Z'-Z$ in (5) must vanish. This can be regarded as a proof of (1).

¹ See, for example, G. Ambrosino and H. Piatier, *Compt. rend.* **232**, 400 (1951). The problem as it affects β -decay has been discussed by Herman M. Schwartz [*Phys. Rev.* **86**, 195 (1952)] with whose final conclusion we concur.

² L. L. Foldy, *Phys. Rev.* **83**, 397 (1951).

³ If one neglects the excitation energy of the excited states of the residual ion, then one can use the completeness relations for the final states of the residual ion and show that the shape of the β -spectrum is not affected, apart from the usual effects of the screened Coulomb field.

Heat Conduction by the Unsaturated Helium II Film

EARL LONG AND LOTHAR MEYER

Institute for the Study of Metals, University of Chicago, Chicago, Illinois

(Received May 21, 1952)

FOLLOWING the recently reported experiments on mass flow in absorbed He II films,¹ preliminary measurements have been made on the transport of heat by such films.

The method used is similar in principle to that of Bowers, Brewer, and Mendelssohn.² A copper-nickel tube 10 cm long, of 0.45-cm i.d. and 0.013-cm wall thickness, had copper chambers attached to top and bottom; the top chamber was provided with a heater; the bottom was in direct contact with the surrounding helium bath. The tube and top chamber were insulated by high vacuum ($<10^{-7}$ mm Hg). Carbon thermometers were located at the center and at both ends of the tube, so that temperature gradients along the tube could be determined. The thermometers were sensitive to 0.0002°.

The heat conductance of the system (without film contribution) was 15 μ watts/degree, so that quite small heat currents could be measured.

The preliminary measurements show:

1. *Saturated films* ($P/P_0=1$, where P is the pressure inside the system and P_0 is the vapor pressure of the bath) show critical heat input rates beyond which the ΔT between top and bottom of the tube rises beyond the experimental range. These critical heat inputs are proportional to the transfer rates of the saturated He II film (Mendelssohn and White), as has already been reported by Bowers *et al.*² However, below the critical heating rate there is always a finite ΔT established for all except quite low heat inputs. The ΔT for a given heat input depends on the amount of excess liquid in the system, but the critical heating rate does not.

2. *Unsaturated films* ($P/P_0 < 1$). For a film of given thickness, determined by P/P_0 , the film contribution to the heat transport is zero ($<0.1 \mu$ watt) at all temperatures above a sharply defined temperature T_f . At this temperature, the film contribution appears abruptly, rising to relatively large values at lower temperatures, depending on the film thickness and the temperature. The temperature T_f is identical with the "onset" temperature for film flow reported by Long and Meyer (reference 1, method I). In Fig. 1

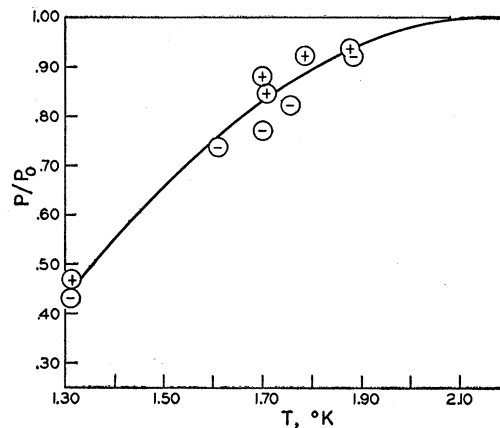


FIG. 1. T_f as function of saturation P/P_0 . (T_f is the temperature above which at a certain saturation, the film does not show superfluidity.)

is shown a curve of these temperatures T_f plotted against the saturation P/P_0 , as derived from the flow measurements of reference 1. The points shown are from the heat transport experiments. For all temperatures above T_f , no film flow and no high heat transport occur; at all temperatures below T_f , film flow occurs and contributes to the heat transport. No attempt was made to accurately re-determine the curve by the heat transport technique, since the flow method previously used is experimentally simpler and more precise.

There is also in the unsaturated films a finite ΔT established in the heat transport experiments. The heat flow is not a linear function of ΔT , but appears to follow roughly $(\Delta T)^{1/2}$, as in the bulk liquid case. The temperature distribution along the tube is such that the ΔT between center and bottom is finite, but much smaller than that between center and top. This suggests that in the heat transport cycle (which presumably involves film flow from cold to warm parts, evaporation, return flow of vapor through the wide tube, then condensation at the cold end) the vaporization process may be the rate-determining step. If, however, the finite ΔT measured in the lower half of the tube is indeed real, then perhaps the film transport cannot be regarded as a true superfluid process.

The heat transport by the film is being investigated in more detail.

¹ E. Long and L. Meyer, *Phys. Rev.* **85**, 1030 (1952).

² Bowers, Brewer, and Mendelssohn, *Phil. Mag.* **42**, 1445 (1951).