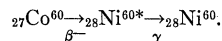


This problem has become of interest in connection with some measurements by Slätis and Siegbahn⁵ on internal pair-formation for determining γ -ray multipole orders in the neighborhood of the threshold. Jaeger and Hulme¹ only made numerical calculations for $Z=0$ and 84 with γ -energies 3, 5, and 7 mc^2 , and consequently their extrapolation to 2 mc^2 in a diagram must be regarded as a little uncertain. Further, the dependence on Z is not easily seen. Therefore, a new numerical calculation has now been carried out with the "exact" theory of Jaeger and Hulme as a basis, where the γ -ray energy 1.31 Mev = 2.57 mc^2 and $Z=28$ has been used—corresponding to the nuclear reaction



(More recent measurements indicate an energy 1.34 Mev = 2.63 mc^2 , but this figure was not known to me when the calculations started.) Jaeger and Hulme presented their results only in a diagram, from which it has not been possible to obtain the actual numbers which they had computed. The figures for $Z=0$ and 84 at γ -ray energies 2.57, 3, 4, and 5 mc^2 have been taken from their diagram, but all these figures are, of course, somewhat uncertain on account of the breadth of the curves and the extrapolation to lower energies. In Table I these figures are presented together with

TABLE I. The internal pair formation coefficient $\times 10^4$. ϵ is the γ -ray energy in mc^2 units. Z is the atomic number of the radiating nucleus. Calculation by Jaeger and Hulme (see reference 1) and the present author.

Dipole case					Quadrupole case						
$Z \setminus \epsilon$	2	2.57	3	4	5	$Z \setminus \epsilon$	2	2.57	3	4	5
0	0	1.40	2.98	6.7 ₀	10.0	0	0	0.38	1.00	3.08	5.6 ₂
28	0	0.91	28	0	0.27
84	0	0.75	1.84	5.3 ₀	8.6 ₀	84	0	0.22	0.76	2.52	4.4 ₂

the results of the new calculations.

The new calculations show that the dependence on Z of the internal pair formation coefficient is not even roughly linear. Within about 10 percent one can, however, get an estimate of the value of the coefficient at different Z in the energy region 2–5 mc^2 by means of the following interpolation formulas. They are rather easy to handle and might be of some use for experimenters.

$$I_D = \exp \left[-\frac{2\pi\alpha Z}{\{2(\epsilon-2)\}^{\frac{1}{2}}} \left(1 - \frac{4.76Z^{\frac{1}{2}}(\epsilon-2)}{1+16(\epsilon-2)} \right) \right] \int_1^{\frac{1}{2}\epsilon+1/\epsilon} \frac{\alpha}{\pi\epsilon^2} \times \frac{dx}{\left(\frac{1}{4}\epsilon^2 + 1 - x\epsilon \right)^{\frac{1}{2}}} \times [2\epsilon(x^2-1)^{\frac{1}{2}} + (\epsilon^2+2-2x\epsilon) \ln \{x+(x^2-1)^{\frac{1}{2}}\}]$$

$$I_Q = \exp \left[-\frac{2\pi\alpha Z}{\{2(\epsilon-2)\}^{\frac{1}{2}}} \left(1 - \frac{2.53Z^{1/8}(\epsilon-2)}{1+4(\epsilon-2)^{5/4}} \right) \right] \int_1^{\frac{1}{2}\epsilon+1/\epsilon} \frac{\alpha}{3\pi\epsilon^4} \times \frac{dx}{\left(\frac{1}{4}\epsilon^2 + 1 - x\epsilon \right)^{\frac{1}{2}}} \times [8\epsilon(x^2-1)^{\frac{1}{2}}(x\epsilon-2) + 3\epsilon^2(\epsilon^2-2x\epsilon) \ln \{x+(x^2-1)^{\frac{1}{2}}\}]$$

where α = fine structure constant = 1/137; Z = atomic number of the γ -radiating nucleus; ϵ = γ -ray energy in mc^2 units; and $x = [E(\epsilon-E)+1]/\epsilon$, where E = total electron energy in mc^2 units. The definite integrals which appear in these expressions are the results of Oppenheimer and Nedelsky for $Z=0$ mentioned above. In Table II the values of the internal pair formation coefficient are computed according to the above formulas in order to make a

TABLE II. The internal pair formation coefficient $\times 10^4$, according to the interpolation formulas. ϵ = the γ -ray in mc^2 units. Z = atomic number of the radiating nucleus.

Dipole case					Quadrupole case						
$Z \setminus \epsilon$	2	2.57	3	4	5	$Z \setminus \epsilon$	2	2.57	3	4	5
0	0	1.46	3.04	6.96	10.4 ₁	0	0	0.35	1.05	3.46	6.06
28	0	0.92	28	0	0.25 ₅
84	0	0.74	2.00	5.45	8.64	84	0	0.20	0.76	2.54	4.34

comparison with the corresponding results of Jaeger and Hulme's theory presented in Table I.

Financial support from the Swedish Atomic Energy Committee is gratefully acknowledged. Further the author wishes to thank Professor Lamek Hulthén and Professor Kai Siegbahn for many valuable discussions.

¹ J. C. Jaeger and H. R. Hulme, Proc. Roy. Soc. (London) **148**, 708 (1935).

² L. Oppenheimer and J. R. Nedelsky, Phys. Rev. **44**, 948 (1933).

³ M. E. Rose, Phys. Rev. **76**, 678 (1949).

⁴ M. E. Rose and G. E. Uhlenbeck, Phys. Rev. **48**, 211 (1935).

⁵ H. Slätis and K. Siegbahn, Ark. Fys. **4**, No. 32, 485 (1952).

Electroluminescence of Single Crystals of ZnS:Cu†

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(Received May 9, 1952)

SINGLE crystals of ZnS:Cu have been prepared¹ and found to luminesce upon application of ac or dc voltages. Ac electroluminescence of powdered sulfide phosphors is well known.² Dc electroluminescence in SiC,³ Ge, and Si⁴ has been studied and attributed to the optical recombination of electrons and holes in an $N-P$ junction. The mechanism of electroluminescence of single crystals of ZnS:Cu is quite different.

With a dc potential applied to a single crystal clamped between metal electrodes, the light output increases exponentially with applied voltage reversible to breakdown. The luminescent intensity is proportional to the current. Different electrode materials alter the brightness-voltage characteristic and permit the identification of the region near the cathode as the origin of the electroluminescence. Pronounced photoconductivity occurs when the cathode side of the crystal is irradiated with ultraviolet.

With an ac potential applied to the single crystal, the light output was determined as a function of the voltage cycle by synchronizing on an oscilloscope the photomultiplier output with the voltage. For low applied voltages two pulses of light per cycle appear out of phase with the voltage (Fig. 1). The pulse width in

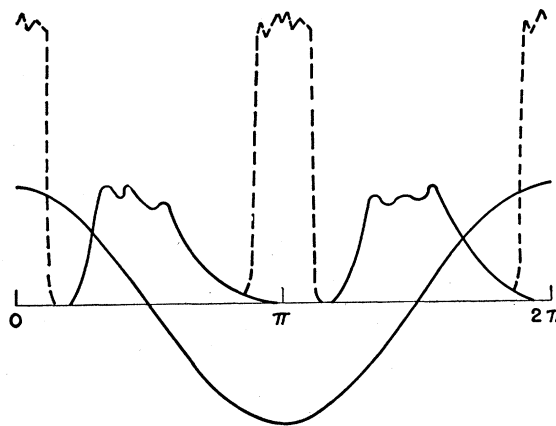


FIG. 1. Voltage and light output versus time.

electrical degrees does not change appreciably from 60 to 6000 cps. The average light output varies linearly with frequency. Each pulse is characteristic of a particular electrode and originates when that electrode is becoming negative during the cycle.

At higher critical voltage, pulses in phase with the applied voltage appear and increase rapidly in intensity with voltage (Fig. 1). Electrodes of high work function such as Pt exhibit a higher threshold voltage for the in-phase component than electrodes of low work function such as Al. With different metals used for the two electrodes, the in-phase pulse typical of a particular electrode material appears when that electrode is negative.

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

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(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 -spectra 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