# Search for a Slow Component in Alpha Ionization

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(Received 5 November 1962)

Delayed ion production has been advanced as the reason for the difference in $W$ (average energy expended in the creation of an ion pair) obtained by fast and slow collection of the ionization from alpha particles in gases. In order to determine whether such slow components exist, the slow and fast measuring methods are applied simultaneously to a single gridded chamber. The results in Ar, 90% Ar+10% $CH_4$, and $N_2$ for alpha energies ranging from 1.5 to 4 MeV show that no such slow components exceeding the uncertainty of the measurements (<0.5%) are present. This indicates that (at least in the above cases) discrepancies between slow and fast methods in past measurements are not due to physical effects.

## I. INTRODUCTION

PAST measurements of $W$ (average energy expended in the creation of an ion pair) for a given alpha-particle energy and gas have, in general, resulted in significantly different values, dependent on the time constant of the measurement ($\sim$a microsecond for electron collection, $\geq$a millisecond for total charge collection). "Fast" measurements gave up to 6% higher values of $W$ than those obtained in "slow" methods.[1] This discrepancy is much higher than the accuracy limits claimed in each method (about 1% or less). The only exception appears to be a recent report by Utterback, Hammer, and Miller.[2] These authors used a fast gridded chamber with a highly purified argon filling. After careful consideration of all corrections, they obtained a $W$ value at the $Po^{210}$ energy which, within the accuracy limit of 1% of the measurement, agreed with the value which Jesse and Sadauskis[3] found by the use of the slow ion collection method.

The dependence of $W_\alpha$ on energy was also investigated by both methods, and the results were conflicting. Jesse and co-workers[4,5] found by the slow method that in the noble gases (particularly in highly purified argon) and in hydrogen $W_\alpha$ was constant for alpha particles ranging in energy from 1 to 9 MeV. Cranshaw and Harvey[6] used 99.8% pure argon in a fast gridded chamber and found that $W_\alpha$ varied with energy according to the (Gerbes type) formula: $W(E)=27.5+1.9E^{-1/2}$ eV/ion pair ($E$ in MeV). The same result was obtained by Harvey *et al.*[7] for an argon plus 6% methane mixture. Since for the 5.3-MeV $Po^{210}$ energy the formula gives $W=28.3$ eV/ion pair, which is 7% higher than Jesse's value of $W=26.4$ eV/ion pair, the formula is now usually quoted in the renormalized[8,5] form $W(E)=W_\infty(1+0.069E^{-1/2})$ retaining only the energy dependence. The formula still implies that $W_\alpha$ at the $Po^{210}$ energy is 3% larger than at very high energies, and that $W_\alpha$ at 1.5 MeV is 2.7% larger than at the $Po^{210}$ energy. Hanna[9] reinvestigated this energy dependence with a gridded ionization chamber which contained argon+2% $BF_3$ and argon+2% $Co_2$ and corroborated the validity of the formula of Cranshaw and Harvey. Similar results were obtained by Rhodes, Franzen, and Stephens.[10]

It is clear that if the production of charges by one alpha particle is confined to a time interval shorter than the time constant applied in the fast method, then both methods should give the same result. A difference in the results seems to imply that there is a slow component in the ion production which is not observed by the fast technique. Such delayed creation of charges can be caused by molecules in excited states (long-lived relative to the time constant of the fast measurement) which ionize trace impurities in the gas. The energy of such excited states also can be reradiated and may release photoelectrons from the chamber walls, as suggested by Hanna.[9] The search for slow components of these types, or of other as yet unidentified types,[11] was the task of the present work.

## II. OUTLINE OF METHOD

The fast and slow measurements are applied simultaneously to the ionization produced by alpha particles in a single gridded chamber. The collector electrode is connected to an electrometer and also (capacitively) to the input of an amplifier system. The number, $N$, of alpha pulses is counted while a total charge, $Q$, is collected. Thus, the average charge $q_\alpha=Q/N$ for one alpha particle is measured by the slow method.[12]

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[8] G. C. Hanna, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1959), Vol. III, Part IX, p. 209.

[9] G. C. Hanna, Phys. Rev. **80**, 530 (1950).

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Simultaneously the pulse-height spectrum of the alpha pulses is taken in a pulse-height analyzer and the average pulse height, $P_\alpha$, is determined (by application of the usual corrections; see below). Then $P_\alpha$ is calibrated in units of charge introduced in the form of a step pulse to the collector electrode. For this purpose, pulses of a nanosecond duration from a pulser are led to the collector electrode through a diode (actually a tetrode of high-internal impedance and negligible backward current). The charge per pulse, $q_c$, is measured in the same way as the charge for one alpha pulse: by the use of the electrometer and by counting; and the average pulse height, $P_c$, for the calibration pulses is obtained from the pulse-height analyzer. Thus $q_\alpha$ and $q_c$ are measured in the same units and $P_\alpha$ is related to $q_\alpha$ in the same way as $P_c$ is to $q_c$.

In order to detect any slow component, $q_\alpha$ and $q_c$ are compared when $P_c=P_\alpha$ (found by linear interpolation between two values, $P_{c1}$, and $P_{c2}$, both close to $P_\alpha$). Any slow component in the ionization process would result in $q_\alpha-q_c>0$.

A collimated alpha beam was used in the measurements to minimize directional dependences in the fast measurement. By changing pressure in the chamber (and hence inside the collimator) the residual alpha energy was changed between 1.5 and 4 MeV. Argon, 90% argon+10% methane, and nitrogen were separately used for the chamber filling, and a recirculating purification system operated during all measurements. We chose these energies and gases in order to treat cases which appeared to be of interest in the controversy mentioned above.

## III. APPARATUS

The gridded chamber in which the ionization measurements were carried out was enclosed by a cylindrical brass housing (128-mm diam and 64 mm high). The collector, grid, and source electrodes were circular disks (84-, 92-, and 107-mm diam, respectively; collector-grid separation 10 mm, grid-source separation 24 mm). A guard ring surrounded the collector; and a skirt (13 mm high and 60 mm diam) extended toward the grid from the source electrode. The housing of the chamber was grounded and the negative voltages $V_g$ and $V_s$ were applied to the grid and source plate, respectively. The convection oven used for gas purification was filled with calcium turnings and operated as described elsewhere.[13] The oven was outgassed between different gas fillings.

The $Po^{210}$ source (a 1-mm-diam spot) was plated on a flat gold surface and mounted inside a cylindrical collimator (11 mm long, 10 mm diam). The collimator window (2 mm×4 mm) was covered by a thin collodion film with gold evaporated over it (total thickness $\sim 5\ \mu g/cm^2$). The collimator was mounted on the source electrode with the alpha beam parallel to the electrode surface.

The collimator has an iron shutter which could be operated by a permanent magnet from outside the chamber. This enabled us to alternate between the alpha, background, and pulser currents.

Pulse calibration was accomplished with a mercury-relay pulse generator and tetrode circuit, shown in Fig. 1. The pulser generated negative pulses of 1-nsec duration at the rate of 60/sec. A shorted line provided an additional pulse of equal height and opposite sign delayed by 1 nsec from the first pulse. This diminished the capacitive feedthrough from cathode to plate of the tetrode. Feedthrough was further diminished (to <0.1% of the main pulse) by the *RC* filter in Fig. 1 ($C_1=2$ pF and two 10 kΩ resistors; the filter had no noticeable effect on alpha pulse heights). The leakage resistances of capacitors $C_1$ and $C_2$ and of the tetrode plate were $>10^{16}\ \Omega$.

The Victoreen VX-41A tetrode was operated with low-interelectrode capacitance by ac grounding its two grids. With 2.6 V negative bias on the grids the quiescent plate current was less than $10^{-15}$ A. A negative

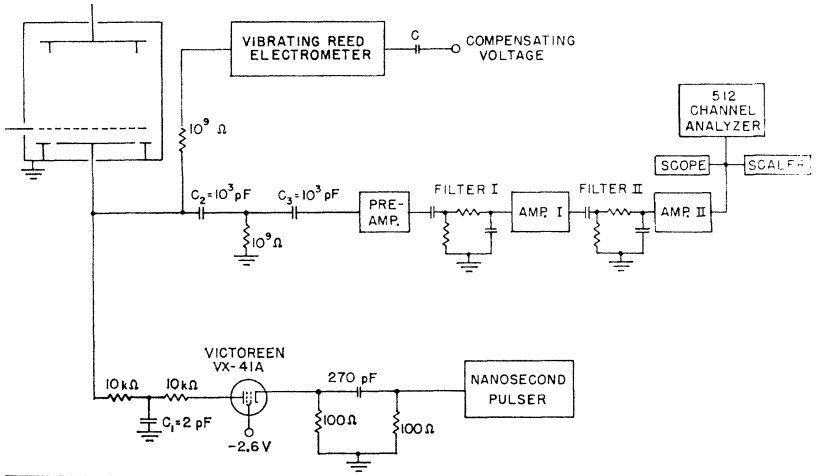


FIG. 1. Block diagram of apparatus.

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swing of about 10 V was required at the cathode to produce about 1 mV at the plate. Thus, the tetrode was insensitive to changes of a few millivolts in its plate circuit caused by charge collection during operation.

The vibrating reed electrometer was used as a null-indicating instrument, in the usual charge-compensating method, for the measurement of charges coming either from the chamber or the tetrode. Fast pulses entered a low-noise preamplifier,[14] were then shaped by two *RC* filtering networks,[12] linearly amplified and passed to a counter and a 512-channel pulse-height analyzer.

A high-pass filter ($C_3=10^3$ pF and $R=10^9$ Ω) prevented slow fluctuations of input grid bias of the preamplifier from introducing spurious charge into the electrometer.

The relays which allowed simultaneous operation of electrometer, scaler, analyzer, and clock are not shown in Fig. 1.

## IV. EXPERIMENTAL PROCEDURES AND CORRECTIONS

Measurements were made alternately with the alpha ionization current and with the pulser current. In each run the total charge $Q$ collected by the electrometer was the same. The value of $Q$ was known to within 0.05%, as was shown by periodic checks of the electrometer capacitance and potentiometer.

Average pulse heights were evaluated as centroids of distributions rather than as peaks or medians. In this way the ratio of the average pulse height to the average charge per pulse (the important quantity in this experiment) was independent of fluctuations in charge entering the collector electrode in successive pulses. Thus, an energy spread in the collimated alpha beam, or a pulse-height spread (even a systematic change) in the output of the pulser during the runs caused no errors. The linearity of the amplifying system and pulse-height analyzer was checked by measuring the slope $dP/dq$ while the output pulse height of the pulser, and therefore $q$, was varied. The slope was found to be constant to better than 1%, while $q$ changed by ±50%. Since the interval of interpolation for $P_c=P_\alpha$ was never larger than about 2% of $P_\alpha$, errors due to nonlinearity were negligible. The time stability of the system was checked by the constancy of $P_\alpha/q_\alpha$ in successive runs. Deviations were found to be <0.2%.

One of the chief advantages of our combined system of a gridded chamber and electrometer is that "saturation" conditions in the chamber can easily be checked. By connecting the grid to the collector electrode and applying high voltage only to the source electrode, the chamber becomes a "two-electrode" chamber in which the presence or absence of saturation is simply established by "charge vs voltage plateau" measurements.

Such experiments showed that in our chamber $q_\alpha$ was constant to within 0.1% while $V_s$ was varied (field strength from ~300 to ~900 V/cm). From this it could be concluded that within those field strength limits charge losses by recombination or charge gains by ion multiplication were negligible in the gases used. The next step was to choose $V_g$ and $V_s$ (in the "three-electrode" chamber) such that $V_s-V_g$ was within the plateau mentioned above, and the measured $q_\alpha$ was the same as in the "two-electrode" chamber. This was always possible in the cases investigated, which also proved that there were no losses of the electronic charge to the grid, housing, or guard ring.

In accordance with Herwig *et al.*,[15] the ratio of the field strength in the grid-collector region to that in the grid-source plate region had to be chosen much higher than the theoretical ratio of one,[16] in most cases higher than three, in order to obtain no dissipation of charges to the grid.

### Correction for Dead Time

The dead time correction was measured by a method described elsewhere,[12] using successively $\tau_1=4$ μsec and $\tau_2=8$ μsec time constants in both Filter I and Filter II shown in Fig. 1. Such counting experiments gave about 0.3% dead time correction to be added to the number obtained with the 8-μsec filters. This result agreed with observation of pulse lengths (24 μsec) on the oscilloscope and counting rates used (about 130/sec).

### Correction for Finite Collection Time

The two time constants $\tau_1=4$ μsec and $\tau_2=8$ μsec were also used to determine this correction. It is well known[8] that this correction is proportional to $S^2/\tau^2$, where $S$ is the collection time. By successive use of the two filters, measuring the difference between the corresponding alpha pulse heights, the correction and $S$ can be determined. Since changing the time constant changes the alpha pulse height by varying the gain of the amplifying system, a reference pulse height with $S=0$ is needed in such experiments. Therefore, step-pulses from the pulser were simultaneously analyzed with the alpha pulses. The results so obtained agreed with estimates of $S$ based on known[13] electron drift velocities in the three gases used.

### Correction for Grid Inefficiency

Following Bunemann *et al.*,[16] we calculated the grid shielding inefficiency, $\sigma$, from $r=0.041$ mm (radius of a grid wire), $d=0.82$ mm (separation of grid wires), and $p=10$ mm (grid-collector separation). We found $\sigma=1.50\%$.

The fractional pulse-height correction due to grid

[14] Oak Ridge National Laboratory Drawing *Q*-2069*A*-2 (by E. Fairstein, unpublished).

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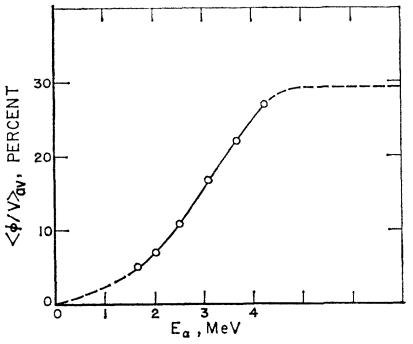


FIG. 2. Measured $\langle\phi/V\rangle_{av}$ vs alpha energy in argon-methane mixture.

inefficiency is then given in a uniform field by

$$\delta P_\alpha/P_\alpha=\sigma(m/a),$$

where $a$ is the grid-source plate separation and $m$ is the distance of a positive point charge from the source plate. For charge distributed along an alpha track, $m$ is the distance from the source plate to the centroid of the charge distribution.

In a nonuniform field the potential averaged over the site of the distribution, $\phi$, must be taken, rather than the geometrical centroid. If $V$ is the total potential difference between source plate and grid, then

$$\delta P/P=\sigma(\phi/V).$$

The ratio $\phi/V$ can be determined by an experimental method. The grid is connected to the collector. With source plate negative, electrons are collected by the grid (+collector), and produce a pulse height

$$P_-=(q/c)(1-\phi/V),$$

where $c$ is the capacity of grid+collector, and $q$ is the total charge. With source plate positive, electrons are collected by the source plate and give a (positive) pulse height

$$P_+=(q/c)(\phi/V).$$

Spectra of both $P_-$ and $P_+$ are taken and the averages $\bar{P}_-$ and $\bar{P}_+$ determined. Then

$$\left\langle\frac{\phi}{V}\right\rangle_{av}=\left\langle\frac{P_+}{P_++P_-}\right\rangle_{av}\sim\frac{\bar{P}_+}{\bar{P}_++\bar{P}_-}.$$

For our collimated alpha beam $\langle\phi/V\rangle_{av}$ vs energy is shown for the argon-methane mixture in Fig. 2. At high energies the curve approaches a limit corresponding to the distance of the beam from the source plate ($m/a\sim7/24$). At intermediate and low energies, the diminishing of $\bar{\phi}$ is caused by the proximity of the charges to the collimator surface.

This experimental method of determining $\langle\phi/V\rangle_{av}$ is useful even in the case of strictly uniform fields, since it eliminates the need for calculating the centroid of the charge distribution from the ionization density. Also, for uncollimated beams the average of $\phi/V$ over the angular distribution is directly obtained.

## V. RESULTS

The results, obtained in the three gases, $N_2$, Ar, and 90% Ar+10% $CH_4$, are tabulated in Table I. In each of these gases, the residual energy of the alpha particles was set by varying the pressure. The different energies ranged between $\sim4$ and $\sim1.5$ MeV. At each pressure the energy of the alpha particles was calculated from the measured $q_\alpha$ and by the values of $W$ as determined by Jesse and collaborators.[3,5] Since the energy values in this work only served as parameters, no estimate of their errors was necessary.

During each $q_\alpha$ run, the charge measured was $\sim5\times10^{-10}$ C which, depending on the energy, corresponded to from $2\times10^4$ to $6\times10^4$ alpha pulses. At each energy in each gas from 3 to 6 such $q_\alpha$ runs were done, and the

TABLE I. Comparison of the ionization charges as measured by the slow and fast methods in argon, 90% argon+10% methane, and nitrogen.

| Gas | Pressure (mm Hg) | Energy (MeV) | $\frac{q_\alpha-q_c}{q_\alpha}$ (%) | Corrections: Dead time (%) | Finite collection time (%) | Grid inefficiency (%) | $\left(\frac{q_\alpha-q_c}{q_\alpha}\right)_{corr}$ (%) | Uncertainty (%) |
|---|---|---|---|---|---|---|---|---|
| $N_2$ | 934 | 4.17 | 1.3 | 0.3 | 0.1 | 0.5 | +0.4 | ±0.4 |
| | 1690 | 2.91 | 1.1 | 0.3 | 0.2 | 0.3 | +0.3 | ±0.4 |
| | 2280 | 1.75 | 0.9 | 0.3 | 0.2 | 0.1 | +0.3 | ±0.4 |
| Ar | 1120 | 3.95 | 1.6 | 0.4 | 0.6 | 0.4 | +0.2 | ±0.4 |
| | 1670 | 3.13 | 1.7 | 0.4 | 0.6 | 0.3 | +0.4 | ±0.4 |
| | 2520 | 1.60 | 1.0 | 0.4 | 0.6 | 0.1 | −0.1 | ±0.4 |
| 90% Ar+10% $CH_4$ | 915 | 4.18 | 0.6 | 0.2 | 0.0 | 0.5 | −0.1 | ±0.2 |
| | 1300 | 3.69 | 0.3 | 0.2 | 0.0 | 0.3 | −0.2 | ±0.3 |
| | 1690 | 3.12 | 0.4 | 0.2 | 0.0 | 0.3 | −0.1 | ±0.3 |
| | 2410 | 1.70 | 0.2 | 0.2 | 0.0 | 0.1 | −0.1 | ±0.3 |

$q_\alpha$ values obtained in each case were constant to within 0.1%. Such accuracy was possible because charge and number of pulses were simultaneously measured.[12]

The measured uncorrected fractional differences $(q_\alpha-q_c)/q_\alpha$ (when $P_\alpha=P_c$) are listed in column 4. The corrections for the dead time, finite collection time and grid inefficiency are given in columns 5, 6, and 7, respectively. Column 8 lists the corrected values of the fractional differences $(q_\alpha-q_c)/q_\alpha$ and represents the results of this experiment.

The uncertainties of the results are listed in the last column. These were compounded from the standard error of the mean and the estimated uncertainties of the corrections and calibrations. The uncertainty of the grid inefficiency correction was estimated to be 30% of its own value due to the uncertainty in the determination of $\sigma$. The uncertainty of the dead-time correction was taken to be 10% of its value due to uncertainties in the knowledge of the filters' time constants. An uncertainty of $\pm 0.1\%$ of the value of $q_\alpha$ was ascribed to the finite-collection-time correction (except in the $Ar+CH_4$ case, where the correction was negligible), because of uncertainties in the knowledge of $P_\alpha$ with two different time constants.

## VI. DISCUSSION

As seen in Table I, none of the corrected fractional differences $(q_\alpha-q_c)/q_\alpha$ exceeds the uncertainty of the measurement. Thus, it is safe to say that, in all cases investigated, the slow and fast measurements give the same result to within somewhat better than one half of one percent. In other words, there is no slow (or delayed) component of the alpha current which is detected in a slow measurement but which remains undetected in fast methods based on electron collection.

It is not the task of the present paper to decide whether in past measurements the results of slow or fast methods should be accepted with greater confidence. The experiences of this work indicate, however, that there are more sources of error inherent in the fast method. Two of these, the calibration of pulse heights in terms of absolute charge and effects of insufficient saturation, were eliminated in the present work. It seems likely that this experiment thereby avoided the main sources of discrepancy between the two methods.

## ACKNOWLEDGMENT

We are indebted to Victor H. Ritz for contributing to the development of the tetrode circuit.

PHYSICAL REVIEW VOLUME 130, NUMBER 1 1 APRIL 1963

# Observations on the Energies of Single-Particle Neutron States*

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(Received 27 November 1962)

The available information on location of neutron single-particle levels is analyzed and the following conclusions are reached: (1) There is strong evidence that the depth of the shell-model potential well varies with symmetry energy, getting shallower as the neutron excess increases. (2) There is a stronger than average interaction between nucleons with the same orbital angular momentum; this causes a level to move down in energy as it fills (self-binding effect), or as a proton state of the same $l$ fills, but the effect seems to be weaker on the $(l-\frac{1}{2})$ neutron state as the $(l+\frac{1}{2})$ neutron state fills. (3) Spin-orbit splittings are extra large when the members of the doublet are in different shells, one full and the other empty; this is attributed to the self-binding effect. (4) The rate of change of binding energy with mass number for a given level, $dE/dA$, is considerably smaller than calculations would indicate; this may be explained as a decrease in potential well depth with $A$, or as a velocity dependence giving an effective mass of nucleons in nuclei somewhat larger than the free nucleon mass. (5) The spacings between oscillator shells is somewhat smaller than in the harmonic oscillator potential, and in available calculations for a Saxon potential; this again may indicate an effective mass greater than the free nucleon mass. (6) The $l$ dependence of the energies of shell-model levels is much smaller than given by Nilsson when the levels are empty, but the Nilsson term gives reasonable agreement when the levels are full; this indicates that the self-binding effect increases with increasing $l$. All of these effects are discussed and quantitative estimates of their magnitudes are given.

## INTRODUCTION

THE location of neutron single-particle states by means of stripping reaction studies has recently been reviewed.[1] The excitation energies from reference 1 are listed in Table I, and the absolute binding energies of ground states from mass and reaction $Q$-value data are listed in Table II. By use of this data, one can readily determine the binding energy of neutrons in the various hole states to the closed-shell nucleus, and the binding energy of neutrons in the various particle states to the closed-shell-plus-one nucleus. A plot of these is given

* Sponsored by the National Science Foundation and the Office of Naval Research.
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