Anomalous Inelastic Scattering of 23-Mev Protons by Heavy Elements

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Measurements of energy distributions from inelastic scattering of 23-Mev protons by heavy elements indicate the presence of very strong, apparently monoenergetic groups. The energies, cross sections, and angular distributions of these proton groups vary slowly and regularly with atomic number. For Z>40, there is little difference between even and odd elements, or across closed shells; this complicates any explanation of these levels by single-particle excitation, and this and other factors make collective-motion explanations difficult.

INTRODUCTION

T is well known that the energy distribution of particles emitted from any nuclear reaction consists of well-defined energy groups corresponding to the various excitation levels of the residual nucleus. In light elements, these groups are well separated in comparison with both their natural width and the resolution of commonly available measuring apparatus, so that reactions such as inelastic proton scattering are commonly used for determining nuclear energy levels. In heavy elements, however, the level spacings become quite small at even 1 Mev of excitation above the ground state, so that one would expect the energy distribution of protons inelastically scattered from such elements, when measured with instrumental resolution wider than these level spacings, to be rather smoothly varying. Furthermore, whatever deviations from smoothness do occur should vary considerably from element to element, reflecting the relatively random spacing of nuclear levels. There should certainly be great differences between even and odd mass elements since their level structures differ markedly.

In the experiments reported here, a very considerable deviation from these expectations was found. The energy distributions of protons inelastically scattered from heavy elements were found to be featured by intense and narrow energy groups for excitations up to at least 4 Mev; there are strong indications that this effect extends up to 10 Mev or higher. Moreover, this structure was found to be quite similar for elements of similar mass, whether odd or even, and the angular distribution of corresponding energy groups in different elements was found to be quite similar. It is this effect that is here referred to as "anomalous inelastic scattering."

EXPERIMENTAL

The experiments were done with the deflected, 23-Mev proton beam of the ORNL 86-inch cyclotron. The beam was collimated, passed through thin targets of the element being studied, and collected in a Faraday cup. The scattered protons were detected by a NaI(Tl) crystal; a 20-channel pulse-height analyzer was used. In some of the measurements, they were first passed through a thin proportional counter, the pulses from which were used to gate the analyzer. This removed a continuum of small pulses from gamma-ray background, but otherwise produced no difference.

To study the performance of the equipment, measurements were made with a natural magnesium target. After correction for center-of-mass motion, the Q value for each energy group corresponded with the energy of a known level of Mg²⁴, the most abundant isotope, within about 5%. These 5% errors were investigated, but no simple instrumental method of eliminating them was found, and they were not sufficiently reproducible from day to day, and as a function of counting rate, to allow the use of a simple correction curve.

The resolution of the detector was such as to give the highest energy groups a full width at half-maximum of about 2.0-2.5% in most of the work reported here; in some of the earlier measurements it was as poor as 3.5%. It was not uniform from day to day, and not insensitive to counting rate. To eliminate the resolution of the scintillation equipment as a factor, and at the same time improve over-all resolution, an absorber was used in front of the scintillator in most measurements. Since the absolute (as opposed to the "percentage") resolution of a scintillator improves as the energy is reduced, and the effective resolution is increased by the



FIG. 1. Demonstration of the absorber-scintillator method. Figure 1(b) shows pulse-height spectrum with no absorber, and Fig. 1(a) shows spectrum with 400 mg/cm² aluminum absorber interposed. The target is cobalt and the detection angle is 90 deg.



FIG. 2. Energy distribution of 23-Mev protons inelastically scattered from elements of atomic number 26–34. The abscissa is actually the pulse height in a NaI(Tl) crystal, but is shown on an approximate energy scale. The ordinate gives relative intensity of points along the individual curves; the curves are displaced arbitrarily in a vertical direction for clarity. The peaks at Q=0 are due to elastically scattered protons. Data for Se are substandard owing to difficulties in target preparation.

ratio of the rate of energy loss at the degraded energy to that at the original energy, the scintillator resolution is readily reduced to about 0.5%. Unfortunately, this resolution is not achieved because energy-loss straggling introduces a resolution spread of about 1.8%. While this does not represent a large improvement over the best resolution achieved with scintillators alone, it frees the experiment from the idiosyncrasies of the latter, improves absolute energy determinations, and spreads the data out so as to give more data points per unit energy. It essentially gives the resolution attainable by differential range detection, but still allows multichannel recording.

Figure 1 shows a pulse-height spectrum from a cobalt target at 90 deg as obtained with a scintillator alone, and with a 400-mg/cm^2 absorber. It is readily seen that the latter has a distinct advantage for quantitative work; it can be obtained with little effort. The former shows a larger portion of the spectrum with fewer data points, but shows less detail; data of this quality can

only be obtained by careful selection of photomultipliers and scintillation crystals, painstaking adjustment of electronic equipment, and use of low counting rates with a steady cyclotron beam. Measurements without the absorber are useful for survey work.

RESULTS

Figures 2–4 show a survey of the pulse-height spectra from various elements. They were obtained with no



FIG. 3. Energy distribution of 23-Mev protons inelastically scattered from elements of atomic number 40–52; see caption for Fig. 2. Mo and Te targets were substandard.

absorber at 90 deg to the incident beam. The abscissa is actually the pulse height, but is shown on a scale which gives approximately the negative Q value of the reaction. The sharp peak at Q=0 corresponds to the elastically scattered protons. The abscissa is not corrected for variations in amplifier gain and cyclotron energy, or for center-of-mass motion, so that there may be 5-10% inaccuracies in this energy scale. In the discussion below, the more correct values are quoted.

Probably the most striking observation from this survey is the strong peak, in most cases obviously double, at $-Q \simeq 2-3$ Mev for all elements between Z=40 and Z=52. The very high intensity and the energy regularity from element to element are very suggestive of an important effect in the nuclear structure of these elements. These groups were investigated in greater detail, as will be discussed below.

For elements in the Z>72 region, the survey (Fig. 4) indicates that the strong groups at $-Q\simeq 2-3$ Mev have disappeared in Ta and W, and appear in Pt and Au with low intensity and large width (which indicates that they are not single groups). The well-known first excited state of Pb²⁰⁸ (Q=-2.6 Mev) is strongly excited, and an equally sharp and intense group is also found in Bi at the same energy. The situation in the -Q=2-3Mev region is somewhat clouded for Th and U by the elastic oxygen peak from oxide contamination (this was indicated by a shift in energy with angle), but there do seem to be broad groups at $-Q\simeq 3$ Mev.

Among the elements with Z = 26-30 (Fig. 2), the proton energy spectrum is featured by very strong groups up to -Q=6 Mev in even-Z elements and up to -Q=4 Mev in odd-Z elements; there is very noticeable structure in the odd-Z elements even up to 8 MeV (see also Fig. 1). Since the spectra for the even and odd elements seem to be somewhat different, they are grouped separately in Fig. 2 rather than being arranged in order of increasing Z; it appears, however, that the main features of the two groups would be reproduced if the energy scales were changed. The principal feature for the odd-Z elements is the two peaks at $-Q \simeq 2.7$ and $-Q \simeq 3.7$ Mev, although the detailed structure of the latter are somewhat different in the two elements. These peaks are markedly similar to the peaks in the even-Z elements at $-Q \simeq 3.1$ and 4.6 MeV, although in this group of elements there is a striking change of the relative intensities of the two peaks between Fe and Ni on the one hand, and Zn on the other. The curve for Se in Fig. 2 should be considered only qualitatively, as the target was thick and nonuniform.

Since practically all of this structure and regularity is most unexpected, a more elaborate investigation of some of the most striking features was undertaken. Careful and detailed measurements were made of the -Q=2-3 Mev groups in the Z=40-52 elements, and to a lesser extent, of the strongest regularly occurring groups in the Z=26-30 elements. Major efforts were made to analyze the spectra into individual groups,



FIG. 4. Energy distribution of 23-Mev protons inelastically scattered from elements of atomic number 73–92; see caption for Fig. 2. Only the Pb and Bi targets were both thin and uniform.

within the limits of the energy resolution of the instruments, to determine their energies accurately (at least relatively among the various elements), and to study their angular distributions. Several independent measurements were made for each element by using the absorber-scintillator method. An example of the energy measurements in the Z=40-52 elements is shown in Fig. 5, and Fig. 1(a) is typical of the data for the Z=26-30 elements.

The results for the Z=40-52 elements are summarized in Fig. 6; these data represent the major effort of this investigation, and give perhaps its most important results. The position of the energy group is plotted vs atomic number; the number of circles in each case gives the differential cross section at 90 deg for that energy group in units of 10^{-28} cm²/sterad. (The angular distribution measurements described below indicate that the total cross sections in milli-



FIG. 5. Pulse-height spectrum from zirconium target with 400-mg/cm² aluminum absorber interposed.

barns are about two times the number of circles.) The open and solid circles represent elements of even and odd atomic number, respectively. In all cases, the observed spectrum up to -Q=3.8 Mev could be completely explained within the experimental resolution by assuming the groups shown are monoenergetic and no other groups are present. The existence of each of the groups was clearly evident with the exception of some of the weak \sim 1.2-Mev groups where, with the exception of In, the situation is typified by Fig. 5. While all data shown were obtained at 90 deg to the incident proton beam, checks were made for each element at 70 deg to ascertain that the energy does not shift with angle as it would if the peaks were due to elastic scattering from light-element impurities. (For example, elastic scattering from an oxide impurity would give an apparent group at -Q=2.3 Mev at 90 deg, but at 1.5 Mev at 70 deg.)

The principal feature of Fig. 6 is the very strong group at $-Q \simeq 2.3$ Mev. For the even-Z elements, the energy shift is small and uniform from element to element except for a slight irregularity (~ 0.2 Mev) at Z = 50. The cross sections are equal within about 20%, which is not much more than the experimental uncertainty. Since all of these elements contain several

isotopes, this indicates that groups of about the same energy must be emitted from each (or at least most) of the isotopes. The odd-Z elements have groups at about the same energy and with the same cross sections. The only apparent difference between these and the groups from the even-Z elements is an energy deviation of about 0.3 Mev for Ag.

Another group occurs in every element at $-Q\simeq 3$ Mev. Here again the energies and cross sections vary quite regularly from element to element, although the energy irregularity at Z=50 is stronger than for the $-Q\simeq 2.3$ Mev group, and there is an additional irregularity of about 0.4 Mev at Z=41. The accuracy of the energy determinations for these groups is poorer than for the $-Q\simeq 2.3$ Mev groups since their cross sections are lower and there is some difficulty in resolving the two groups. There is a cross-section irregularity between Z=50 and Z=52.

An additional group is observed in all odd-Z elements and about half of the even-Z elements at $-Q \simeq 1.2$ Mev. The energy is quite regular for the odd-Z elements, but the cross section varies somewhat, especially for Z=49(In) where it is very strong. This group would have been detected in Pd and Cd if it were one-fourth as strong as in Sn, so this again may be considered as a cross-section irregularity. There is a considerable uncertainty in the energy determinations for these groups (except for In) due to difficulty in resolving them from the elastically scattered protons.



FIG. 6. Energy levels in Z=40-52 elements required to explain inelastic proton scattering spectrum. Open circles are for even-Z elements, filled circles for odd-Z elements. The number of circles gives the differential cross section at 90 deg in units of 10^{-28} cm²/sterad. Triangle indicates $\sigma \simeq \frac{1}{2} \times 10^{-28}$ cm²/std.

The data for the Z=26-30 elements are summarized in Fig. 7, where the representation is identical with that of Fig. 6. Here the $-Q\simeq 1.2$ Mev levels which are so strongly excited are the well-known first excited states. For the even-Z elements, the energies and cross sections are quite regular except for the greatly increased cross section for the -Q=3.0 Mev group in Zn, and the weak extra and missing groups in Fe at -Q=2.8 and 4.4 Mev, respectively. The latter irregularities can be explained by lack of resolution, but the intense excitation of the Zn level is very clearly evident (see Fig. 2).

The Co and Cu data are quite similar to each other except for a factor of six cross-section irregularity in the $-Q\simeq 2.1$ Mev groups. The relationship between the even- and odd-Z elements is not clear but the energy trends seem to be parallel.

The greatest effort on angular distributions was concentrated on the $-Q \simeq 2.3$ Mev groups in the Z=40-52 elements, and the $-Q \simeq 3.1$ Mev groups in the even-Z =26-30 elements; the data are shown in Fig. 8. At angles smaller than \sim 35 deg, the groups in question could not be resolved from the continuum. There was also some difficulty of this type in the region of the minimum at ~ 65 deg. In the backward direction, the resolution was somewhat poorer because of target thickness. For Zr, Ag, and Sn, the angular distribution of the $-Q \simeq 3$ Mev groups was apparently quite similar to those shown in Fig. 8, although they were not determined with very good accuracy except for Zr. In that element, the maximum at 40 deg seemed to be shifted to about 43 deg, and the curve in this region seemed to fall off more steeply at smaller angles and less steeply at larger angles than the lower energy group. The angular distribution for the -Q=3.3 MeV group in Fe seemed quite similar to the corresponding



FIG. 7. Energy levels in Z=26-30 elements required to explain inelastic proton scattering spectrum; see caption for Fig. 6.



FIG. 8. Angular distributions of $-Q \simeq 2.3$ Mev groups in Z = 40-52 elements and of $-Q \simeq 3.0$ Mev groups in Z = 26-30 elements.

group in Ni, although the accuracy of measurement was somewhat poorer.

In accordance with the theoretical work of Austern, Butler, and McManus,¹ angular distributions $I(\theta)$ from direct interactions should be represented by

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$$V(\theta) = \sum_{l} C_{l} J_{l}^{2} ([K_{i} - K_{0}]r), \qquad (1)$$

where K_i and K_0 are the wave numbers of the incident and outgoing protons, r is the interaction radius, presumably about equal to the nuclear radius, J_l are the Bessel functions of order l, C_l are undetermined constants, and the summation is over values of l which can be reached by a vector sum of the spins of the initial and final nuclei and the incident and outgoing proton. Only even (odd) values of l are included if the parities of the initial and final nuclei are the same (opposite).

The principal feature of Eq. (1) that is expected to fit the data is the position of the first (and possibly the second) maximum. The comparison with the data of Fig. 8 is shown in Fig. 9 for the $-Q\simeq 2.3$ Mev groups in the Z=40-52 elements and in Fig. 10 for the $-Q\simeq 3.1$ Mev groups in the Z=26-30 elements. In these figures, curves are shown for $(K_i-K_0)r$ for various values of rrepresented by the usual formula

$$r = r_0 A^{\frac{1}{2}} \times 10^{-13} \,\mathrm{cm}.$$
 (2)

In addition, the values of the ordinate for which J_l^2 has a maximum are shown for each l at the positions of the observed maxima in the angular distributions. For a

¹ Austern, Butler, and McManus, Phys. Rev. 92, 350 (1953).



FIG. 9. Comparison of maxima in angular distributions with theoretical predictions for Z=40-52 elements.

consistent fit to be obtained, the curve of $(K_i - K_0)r$ for a given radius should pass through lines for a given l(or for an *l* larger by 2 units, 4 units, etc., in some cases) at the position of each observed maximum. From Fig. 9, it is seen that a fit can be obtained for l=0 at $r_0=1.2$, for l=3 at $r_0=1.4$, for l=4 at $r_0=1.6$, and possibly for l=1 at $r_0=1.55$. A fit for l=2 would require $r_0=1.0$, a fit for l=5 would require $r_0=1.9$, and no reasonable fit can be obtained for higher values of l. From Fig. 10 it is seen that the same values of l for the same r_0 's are obtained. This is due to the fact that the positions of the two observed maxima occur at the same values of $(K_i - K_0)r$ in spite of the fact that the angles at which they occur, the values of K_0 (i.e., the *Q* of the reactions), and the nuclear radii are different. This strongly suggests that the $-Q \simeq 3.0$ groups in Z=26-30 elements arise from the same source as the $-Q \simeq 2.3$ MeV groups in the Z = 40-52 elements. This is further evidenced by the fact that the energy trends within each of the two mass regions are in the correct direction and of approximately the correct magnitude; in addition it may be noted that the rough data for Se (Z=34) indicates a strong peak at $-0 \simeq 2.6$ Mev.

The very deep minimum in the Ni (and also Fe) angular distribution suggests that this may be due to a zero of the Bessel function. These zeros are therefore shown in Fig. 10 at the angle at which the observed minimum occurs. It is seen that the fit is satisfactory for any of the possibilities mentioned above.

In addition to the measurements shown in Fig. 8, rough data were obtained for several other groups. In all cases, the angular distributions were strongly forward, and in general their increase in the forward direction was more rapid than for the curves of Fig. 8. In cases where well-defined groups were studied, there were indications of minima and secondary maxima. The



FIG. 10. Comparison of maxima and minima in angular distributions with theoretical predictions for Z=26-30 elements.

-Q=2.6 Mev groups in Pb and Bi have very similar angular distributions.

CONCLUSIONS AND DISCUSSION

A. Implications for Nuclear Reaction Theory

From the standpoint of nuclear reaction theory, the most interesting conclusion from this data is that transition probabilities to different nuclear levels can vary by many orders of magnitude. For example, from Fig. 3, the area under the $-Q\simeq 2.3$ Mev groups in the Z=40-52 elements are of the same order as the area under a 1-Mev section of the spectrum in the region $-Q\simeq 8$ Mev. In the latter region, the level densities are known from neutron capture data to be of the order of 10^5 per Mev; thus, one must conclude that the average transition probability to those levels is smaller by a factor of 10^5 than that to the $-Q\simeq 2.3$ Mev level.

As an extension of this argument, it might be noted that if there are levels at $-Q\simeq 2.3$ Mev which can be excited so strongly, there is no obvious reason why some levels at $-Q\simeq 8$ Mev cannot be excited with strengths of the same order of magnitude. This would imply that even at high excitation energies only a few levels per Mev are excited with appreciable strength. This would then explain the irregularities observed in the lowenergy (i.e., large values of -Q) portion of the spectra in Figs. 2-4. These irregularities would not occur even if a hundred levels per Mev were excited, unless, of course, there were nonstatistical regularities in the spacings and/or transition probabilities. Such regularities could result from "giant resonance effects."²

It appears from the angular distribution data that all reactions being studied here proceed by a "direct" rather than by a "compound-nucleus" interaction. This includes the intense, broad peak at $-Q \simeq 7$ Mev in the

² Lane, Thomas, and Wigner, Phys. Rev. 98, 693 (1955).

heavy elements, which have cross sections of the order of 100 mb, so that direct-interaction cross sections must be at least that large. This is consistent with the results of Eisberg and Igo³ who found total (p, p') cross sections ~ 200 mb for 32-Mev bombarding energy.

B. Implications for Nuclear Structure

In considering the nuclear structure problems raised by the results of this experiment, attention is given especially to the -Q=2-3 Mev groups in the Z=40-52elements since these were investigated most thoroughly. The regularities in the energies and cross sections as exhibited in Fig. 6, and the similarities of the angular distributions of Fig. 8 indicate quite positively that there is a strong relationship between the corresponding levels in the different elements. Since these levels are excited by a direct interaction, they evidently have a high fractional parentage coefficient with the ground state. Two general types of levels are usually considered to have such a high fractional parentage coefficient, namely, those arising from collective motions and from single-particle excitation.

The excitation energies involved are generally more typical of those usually considered for single-particle excitation. Moreover, the lower of the two levels in Zr is known from beta decay as a level of Zr⁹⁰, and there is very good evidence⁴ that it arises from single-particle excitation. It is, of course, possible that the observed peak is due to the other isotopes of that element (totalling 48.5%) but this would introduce an irregularity into the cross section data of Fig. 6. One additional evidence of this type is the case of Pb²⁰⁸ where the first excited state, -Q=2.6 Mev, is strongly excited (see Fig. 4). It does not seem implausible to connect this state with the $-Q \simeq 2.3$ Mev levels in the Z = 40-52elements. There is very good evidence⁵ that the Pb^{208} state is due to single-particle excitation. On the other hand, the strong similarity between this level in Pb and the one at the same energy in Bi is extremely puzzling; the accepted explanation for the Pb level, namely single proton excitation, would certainly not explain the Bi level. The rather remarkable correspondence between the spectra and angular distributions for Pb and Bi is under further investigation.

The principal objection to the single-particle excitation explanation is the regularity between even and odd elements and across closed shells. In an even-Z

element (these consist principally of even-even isotopes), a single-particle excitation involves breakup of a nucleon pair, whereas in an odd-Z nucleus it does not, so that the single-particle excitation energy should be much less. Furthermore, one would expect large irregularities just beyond Zr whose most abundant isotope has 50 neutrons (and 40 protons), and just beyond Sn which has 50 protons. Neither of these irregularities is strongly in evidence, and there is also no irregularity in the region of 28 protons (Ni).

A collective-motion explanation might avoid the difficulty from the similarity between odd and even nuclei, although it would still be difficult to see why there should not be large effects at closed shells. However, the energies are considerably higher than generally expected from collective oscillations6; they are much higher than the levels investigated by Scharff-Goldhaber and Weneser,⁷ which were found to have all the properties expected of collective oscillations, such as uniform level spacing, correct spins and parities, large cross sections for Coulomb excitation, and decay by E2 transitions far more rapid than expected for singleparticle transitions. Another difficulty with a collective oscillation explanation of the levels studied here is that the value l=2 seems to be excluded by the analysis of the angular distributions; this is the value expected for the lowest lying state arising from collective oscillations.

It thus seems difficult to reconcile the evidence on the levels observed here with the expected properties of either of the two types of levels that are generally considered to have a large fractional parentage coefficient with the ground state. However, the properties of these levels are extremely suggestive, and it seems quite certain that their explanation will throw important light on problems of nuclear structure. When such an explanation has been achieved, anomalous inelastic proton scattering may well provide an important tool for further investigations.

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³ R. M. Eisberg and G. Igo, Phys. Rev. 94, 739 (1954).

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⁶A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. ⁷G. Scharff-Goldhaber and J. Weneser, Phys. Rev. 98, 212

^{(1955).}