Fluorescent Response of NaI(Tl) and CsI(Tl) Crystals to Heavy Ions of Energies 1 to 300 keV*

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The relative fluorescence output of freshly cleaved NaI(Tl) single crystals under bombardment with ions of mass from 1 to 133 amu was determined as a function of particle kinetic energy, in keV, as follows: H and D from 1 to 100; He from 10 to 100; Li from 20 to 100; C from 10 to 200; N from 10 to 200; O, F, and Na from 20 to 200; Ne from 15 to 200; Al from 30 to 100; Ar and Xe from 30 to 200; I from 50 to 100; Kr from 60 to 200; and Cs from 50 to 200. Except for H, D, Ne, and Na ions, the pulse-height–versus–energy relation for each different-mass ion shows two distinct linear regions. The change in slope for ions of mass less than that of Na is negative; for ions of mass greater than that of Na the change in slope is positive. In the energy range used, the slope of the pulse-height–versus–energy relation decreases as the incident ion mass is increased. In CsI(Tl) the pulse-height–versus–energy relation is linear for all the different ions used. The ratio of pulse height from NaI(Tl) to that from CsI(Tl) for a given mass and energy ion was typically 2.3. However, because of the slope change for NaI(Tl) the ratio varied from 2.0 to 2.5. Except for a few unexplained instances, the resolution by NaI(Tl) is approximately proportional to $E^{-1/2}$. A theoretical calculation based on a model of energy transfer by the incident particles to the principal constituents of the crystal lattice is presented. In this model, the energy transfer is divided into light-producing atom-electron interactions and atom-atom interactions in which negligible light production occurs. The model can explain logically the decreasing scintillation efficiency at a given energy as the incident particle mass is increased. Theories of Bohr and of Lindhard, Scharff, and Schiott are coupled in the development of the model used. Universal pulse-height–versus–energy relations result.

I. INTRODUCTION

IN the open literature only a small amount of research has been reported in connection with the response of scintillation crystals upon bombardment with heavy ions of kinetic energy in the region of 100 keV and less. In a recent paper[1] by the present authors work has been reported in which this type of crystal was bombarded with ions of mass from 1–200 amu with kinetic energy up to 300 keV and a measure of their relative fluorescence output given. Substantial information is available[2,3] about the response of these crystals to heavy ions up to fission fragments; however, in general the particle energies have been from the low-MeV region up to around 160 MeV. In this paper, work is reported in which NaI(Tl) was bombarded with ions in the mass range from 1–133 amu with kinetic energy up to 200 keV and a measure of their relative fluorescence output given. Also included is a theoretical interpretation of the results in the previous paper[1] and of the NaI(Tl) results.

Aside from the practical applications, knowledge of the response of scintillation crystals to low-energy heavy ions is of value in testing and developing theories which lead to an understanding of the scintillation and energy-loss processes in these crystals.

For this investigation, NaI(Tl) and CsI(Tl) were chosen mainly because these are used more often than other inorganic crystals in experiments wherein the scintillation process is involved. Because the scintillation response is mass- and energy-dependent but nearly charge independent for low-charge states of the incident ion, such a detector system is particularly useful for the detection and identification of low-keV-energy neutral particles. Of the two, CsI(Tl) is easier to use because of its nonhygroscopicity and, thus, reduced probability of formation of an energy-degrading surface layer. Also, of the commonly used nonhygroscopic single crystal scintillators it gives the largest light output for the heavy ions. In contrast, NaI(Tl) is very hygroscopic. However, of the inorganic single crystals presently in use it has the greatest light yield for a given ion of given energy. The surface characteristics of this material can change very easily; this fact lead to a series of experiments briefly described later.

Past work[2–9] has shown that for the heavy ions, the maximum scintillation efficiency from the crystals of interest in this investigation occurs for protons and deuterons. For these particles into NaI(Tl) the relationship between pulse height (PH) and energy (E) is linear within experimental uncertainty for all energies from 4 keV to 20 MeV; this is not true for CsI(Tl).[10] Also, the scintillation efficiency at a given energy decreases, and the light response versus energy becomes nonlinear in the MeV-energy region as the incident ion mass is increased.

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[1] G. J. Lockwood and G. L. Cano, IEEE Trans. Nucl. Sci. NS-13, 716 (1966).
[2] A. R. Quinton, C. E. Anderson, and W. J. Knox, Phys. Rev. 115, 886 (1959).
[3] C. B. Fulmer, Phys. Rev. 108, 1113 (1957).
[4] E. Newman, A. M. Smith, and F. E. Steigert, Phys. Rev. 122, 1520 (1961).
[5] S. K. Allison and H. Casson, Phys. Rev. 90, 880 (1953).
[6] R. W. Hill, Rev. Sci. Instr. 33, 1477 (1962).
[7] S. Bashkin, R. R. Carlson, R. A. Douglas, and J. A. Jacobs, Phys. Rev. 109, 434 (1958).
[8] F. S. Eby and W. K. Jentschke, Phys. Rev. 96, 911 (1954).
[9] R. B. Murray and A. Meyer, Phys. Rev. 122, 815 (1961).
[10] R. Gwin and R. B. Murray, Phys. Rev. 131, 501 (1963).

For NaI(Tl) the present results support and, in energy and mass coverage, extend past findings. In the low-energy region of this experiment, the PH versus E relation is linear for all the different mass ions used, but a change in slope occurs in most cases. This nonlinearity may have a physical cause distinct from that of the nonlinearity which occurs at very high energies. For CsI(Tl) it was found[1] that the efficiency decreased rapidly as mass was increased, but the relationship between light output and energy for energy from 1–300 keV was linear within experimental uncertainty. This is not inconsistent with past work[2,4,10] as the present curves from 1 keV to less than 300 keV, in most cases, are but a small addition to the curves which extend from less than 1–160 MeV. The latter curves show the nonlinearity, which occurs in the several-MeV region, quite clearly.

It was shown in a number of papers[8,9] that for a given scintillator the nonlinear relationship between PH and E which resulted when the crystal was bombarded with high-mass atomic particles was due possibly to a saturation of the light-producing centers of the crystal; i.e., at some critical value of stopping power, $(dE/dx)_c$, enough of the light-producing centers along some portion of the path and available to the energy carriers were utilized so that if dE/dx exceeded $(dE/dx)_c$, the slope of the PH versus E relation decreased. The additional energy loss was to the lattice by some processes which did not yield light. The decrease in PH which resulted at a given energy, in the MeV region, as ion mass was increased was explained as being due to a greater dE/dx value as the ion mass was increased. Finally, it was concluded[8,10] on the basis of experimental evidence that the activator saturation mechanism was not responsible for the decline of scintillation efficiency at large dE/dx. The decrease in efficiency was attributed to some intrinsic property of the host crystal and was relatively independent of activator concentration.

For the low-energy, high-mass region of this experiment, an alternative theory to the saturation theory is presented in this paper. This alternative theory is based on nuclear collisions[11] in which momentum and kinetic energy are transferred by the incident atom to translatory motion of the target atom as a whole, and on electronic collisions[11] in which kinetic energy is transferred to the individual electrons of the target atoms resulting in atomic excitation and ionization. The decreasing scintillation efficiency at a given energy as the incident particle mass is increased is explained by this theory.

At a given energy the dE/dx for a given scintillator increases as the incident particle mass is increased.[8,9] Now, if at the given energy the increased dE/dx in the crystal as the incoming particle mass is increased is mainly through elastic and inelastic processes in which negligible light production occurs, then at the given energy the PH should decrease as ion mass is increased. It is important to emphasize that this decrease in light output per unit energy input may not be due to a saturation of light-producing centers but alternatively that it could be due to a large amount of energy loss through primary interactions in which light is produced with negligible efficiency. In the low-energy region of this experiment, the interactions necessarily[11] would be nuclear collisions in which the incident ion transfers kinetic energy to the resident ions of the crystal lattice. The recoil lattice ions on the average would attain a velocity of from one to two orders of magnitude less than the Bohr velocity, $V_0=2.19\times10^8$ cm/sec. Because of this they would lose a significantly greater fraction of their kinetic energy in further nuclear collisions than the primary incident particle. Thus, only a small fraction of the total light output would be due to the recoil atoms.

To examine the hypotheses above, the amount of energy lost by the incoming particle elastically in nuclear collisions on the one hand, and inelastically in electronic collisions on the other was calculated. The two types of collisions are not mutually exclusive in nature, but for mathematical simplicity, it is so assumed. Then from the theoretical determination of the separate contributions, universal curves of PH versus E for a given scintillator were obtained. That is, if the measured pulse height is plotted, not against particle initial kinetic energy, but rather against the calculated energy loss to electronic inelastic collisions, then the PH versus E lines of all particles fall into two or three distinct regions. A detailed explanation is given in Sec. V.

II. APPARATUS AND EXPERIMENTAL PROCEDURE

The heavy ions used in these measurements were from an rf ion source[12] of the Oak Ridge type which is part of the Sandia Laboratory 100-kV ion accelerator.[13] By magnetic selection of triply ionized particles the useful range was extended to 300 keV in two cases. The particle energy was known to within 2%.

In Table I are listed the 16 ions and corresponding energy range used on NaI(Tl). In Table II, for comparison, are listed the 11 ions and energy range used on CsI(Tl).

For this study the beam of accelerated particles was collimated by two 0.44-mm diameter circular apertures 23 cm apart. The intensity of this primary collimated beam was approximately 10^{-9} A. A beam of singly ionized particles of this intensity corresponds to approximately 6×10^9 particles per second. This many particles incident on a 0.44-mm diameter circular area of the crystal either would have damaged that area or

[11] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. Phys. Medd 18, No. 8, 1 (1948).

[12] G. J. Lockwood, Rev. Sci. Instr. 37, 226 (1966).

[13] Obtained from Texas Nuclear Corporation.

TABLE I. Ions incident on NaI(Tl) and their corresponding energy range.

Ion	Energy range (keV)	Ion	Energy range (keV)	Ion	Energy range (keV)
H	1–100	O	20–200	Kr	60–200
D	1–100	F	20–200	I	50–100
He	10–100	Ne	15–200	Xe	30–200
Li	20–100	Na	20–200	Cs	50–200
C	10–200	Al	30–100		
N	10–200	Ar	30–200		

TABLE II. Ions incident on CsI(Tl) and their corresponding energy range.

Ion	Energy range (keV)	Ion	Energy range (keV)	Ion	Energy range (keV)
H	1–100	N	40–200	Kr	60–200
D	1–100	O	20–200	Xe	70–200
He	10–100	Ne	30–200	Hg	100–300
C	16–300	Ar	60–200		

would have overloaded the associated electronics, or both, depending on the particle energy. Thus, in order to do pulse-height analysis, only a small fraction of the primary beam of particles was utilized. Two methods were tried to reduce the intensity of the particle beam which struck the crystal. The primary beam was defocused in one case and allowed to scatter from a target gas in the other case before impinging on the crystal. No difference was observed between the resultant PH distributions at a given energy. Thus, because the scattered beam was more readily available, the scattered beam method was utilized.

Figure 1 shows the scattering chamber and detecting system used. The collimated primary ion beam entered the scattering chamber through hole A. A movable monitor behind hole A allowed the primary beam to be selected and stabilized before scattering took place. The primary beam was then allowed to scatter from the residual gas in the region between holes A and B. The detecting system was pivoted about the center of the scattering region to the desired angle, θ. The scattered beam was defined by two 1.0-mm-diam circular apertures 6.4 cm apart, B and C, and then passed between a set of electrostatic analyzer plates before reaching the detector system.

The detector system consisted of the crystal[14] being investigated mounted directly on the end-window of a Dumont 6292 photomultiplier tube (PMT) taken from stock. Optical contact between crystal and PMT was accomplished by means of 10^4-centistoke silicone oil. The PMT end-window formed part of the vacuum wall and, thus, the crystal was inside the vacuum chamber exposed directly to the scattered particle beam. The particles struck the crystal at normal incidence near its center after passing through a small hole in a light-reflecting well which housed the crystal and PMT face.

The decay time of NaI(Tl) is approximately 0.3 microsecond and that of CsI(Tl) is approximately 1.1 μsec. The transit time of a pulse through the PMT was about 0.03 μsec. The output pulses from the PMT were fed undistorted through an RC circuit of time constant 1.0 msec to a cathode follower and then to a conventional low-noise, high-gain linear amplifier. After amplification the signal pulses were processed in a 256-channel pulse-height analyzer.

In an attempt to determine what differences occurred in pulse height, pulse shape, and resolution for a given ion species as a function of crystal surface condition, three different sets of experiments were performed with NaI(Tl). In one, a polished crystal of unknown axial orientation was used as obtained from the factory. In another set, a 0.050-in. thick crystal was used which had been freshly cleaved along the commonly obtained 001 plane and coupled to the PMT in a dry argon atmosphere and then transferred through 40% relative humidity air to the accelerator port from which dry argon was effusing. In the third set a crystal was used which had been freshly cleaved along the 001 plane and

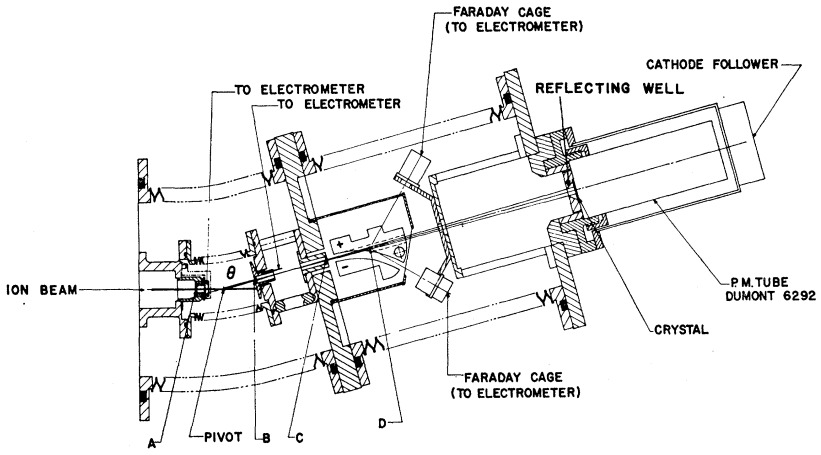

FIG. 1. Experimental apparatus. A is the scattering chamber 0.44-mm-diam entrance aperture. θ is the angle of scatter for the particle beam defined by the 1.0-mm-diam apertures B and C. D defines the virtual axis of the analyzer plates.

[14] Obtained from the Harshaw Chemical Company.

exposed only to an inert atmosphere. The pressure in the test chamber was reduced to $\lesssim 10^{-7}$ Torr within 15 min after crystal installation. The cleavage on both sides of the crystals was along a cleavage plane as seen by the eye. The CsI(Tl) crystals were used as obtained. Both CsI and NaI had a nominal thallium content of 0.1 mole %.

In obtaining the PH distributions the scattering angle θ was adjusted so that the counts per channel near the peak of the PH distribution numbered between 1000 and 50 000 for an average bombarding time of one minute. Pulse heights and resolution were obtained from plots made from the typed output of the analyzer. Corrections were made for background noise.

The use of multiply ionized particles to extend the useful range of the accelerator was justified as follows. The scattered beam was a composite of neutral and different charge state particles. Because of this a set of measurements was made to determine whether or not there was a significant difference in the pulse height distributions obtained from the crystal for incident neutral particles and for particles with charge from $+1e$ to $+5e$ of a given energy. For these measurements the detector system was rotated about the virtual axis, D, Fig. 1, of the analyzer plates to a position where the particles of a given charge could be swept to the crystal one charge number at a time. No difference was observed in the PH distributions obtained for Al or the Al^{+1} to Al^{+5} ions of a given energy in our low-energy region. Further, in a comparison of the PH for doubly ionized particles with the PH for singly ionized particles of the same isotope which had been accelerated through twice the potential as the doubly ionized particles no difference was observed for any of the ten different mass ions for which doubly ionized particles were used.

The stability of the crystal electronics system was checked throughout the experiment by bombarding the NaI(Tl) crystal with 100-keV protons after each different mass ion was used. All 100-keV proton pulse heights were the same, within experimental uncertainty, from beginning to end of the experiment.

III. RESULTS

Results of the measurements performed using CsI(Tl) have been given elsewhere.[1] For convenience, the PH versus E curves are reproduced in Fig. 6, however. Results of the measurements performed using NaI(Tl) are shown in Figs. 2 and 3. Figure 2 is a plot of relative pulse height versus particle energy and Fig. 3 illustrates the resolution, defined as the ratio of the full width of the peak at half-maximum intensity to the peak central value, in percent as a function of energy. The linear least-squares fit to the data is shown by solid lines. Within experimental uncertainty, this linear fitting process was appropriate. The unit of relative pulse height for all the data presented was fixed

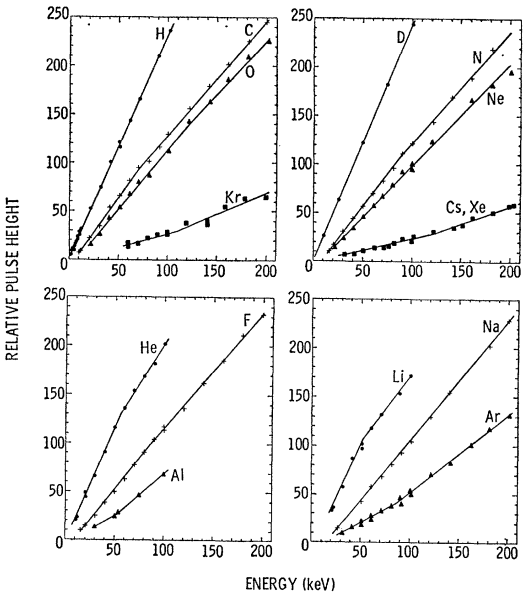

FIG. 2. Relative pulse height from NaI(Tl) versus particle energy for a number of heavy ions. Points are experimental. Curves are least-squares fits to the data.

by setting the PH for the 100 keV H^+ ion to 100 for CsI(Tl).

A composite picture of the linear least squares fit to the PH versus E data for each type of particle is given in Fig. 4. Figure 5 is a similar composite plot of the linear least-squares fit to the resolution data.

Figure 7 shows a plot of PH versus E for H and Kr in the two crystals. This illustrates clearly the ratio of PH from freshly cleaved NaI(Tl) to that from polished CsI(Tl) when bombarded by ions of a given mass. A ratio of ~ 2.3 was common.

The results of the investigation on possible differences in response of the three differently obtained NaI(Tl)

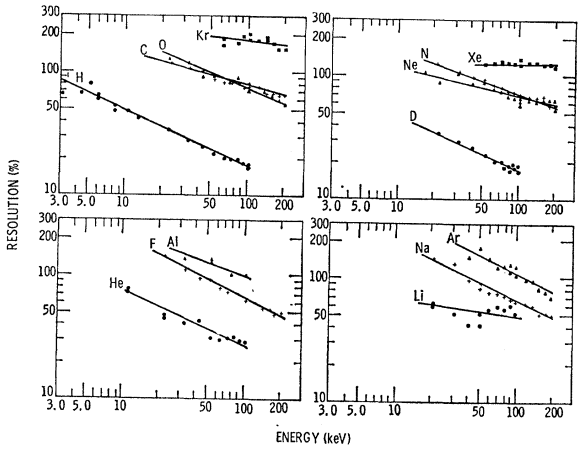

FIG. 3. Resolution of pulse-height distribution by NaI(Tl) versus particle energy for a number of heavy ions. Points are experimental. Curves are least-squares fits to the data.

crystal surfaces showed distinct effects. The pulse-height distributions from the polished crystal for all the ions used were asymmetric. This may be due to energy loss by the incident ions to microscopic particles left on the crystal surface from the final polishing rouge. The crystal cleaved in an inert gas but transferred in air showed the same effects as the polished crystal but to less extent. The freshly cleaved crystal which was exposed only to inert dry gases yielded the highest PH for a given energy ion, and for the low mass ions the pulse-height distributions were symmetric. Therefore, only cleaved crystals subjected to an inert atmosphere were used in obtaining the results reported herein.

IV. EVALUATION

It is seen in Fig. 2 that other than for H, D, Ne, and Na, the PH versus E relation for each of the different mass ions shows two distinct linear regions with the change in slope occurring at a greater value of energy as the ion mass is increased. Note that, as energy increases, the slope change is less than zero for the ions of mass less than that of Na, and greater than zero for the ions of mass greater than that of Na. The phenomenon of slope change is not observed with CsI(Tl), Fig. 6. It is not known why the difference arises. The following observations are made, however. The mass of Na is substantially different from that of I; Cs and I are almost the same. Also, if the slope change for each PH versus E curve were due either to a different cell structure (NaI: fcc; CsI: bcc) or lattice spacing (NaI: 6.48 Å; CsI: 4.57 Å), then one would expect the break to occur systematically in NaI(Tl) for all the ions that were not completely stripped of electrons, regardless of mass.

The slope change is greatest for those ions whose mass is substantially different from that of Na. H and D are anomalous in this respect, but this can be explained as follows. A proton of energy ≥ 25 keV has a velocity, $V \geq V_0$. The same is true of deuterons of $E \geq 50$ keV. Therefore, these particles are stripped of their electron for a considerable part of their range in the crystal. If the pronounced change in slope is due to an atom-atom type of interaction, the change in slope might then be absent for a nucleus-atom type of interaction. No explanation is offered as to why either a change in slope occurs for most of the ions used or no change occurs for Na and Ne.

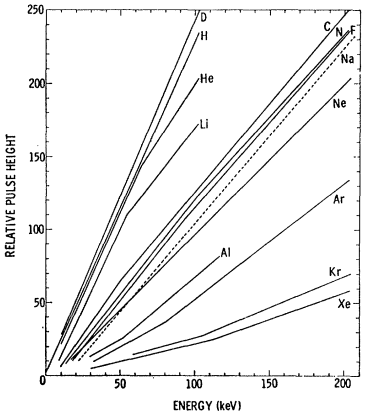

FIG. 4. Least-squares curves of relative pulse height from NaI(Tl) versus particle energy. A composite of the separate curves is shown for comparison.

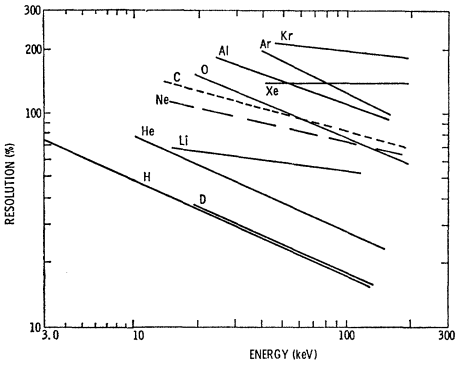

FIG. 5. Least-squares curves of resolution by NaI(Tl) versus particle energy. A composite of the separate curves.

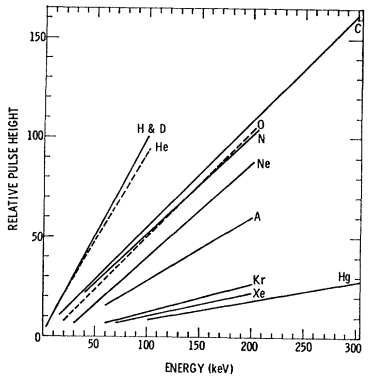

FIG. 6. Least-squares curves of relative pulse height from CsI(Tl) versus particle energy. A composite of the separate curves (Ref. 1).

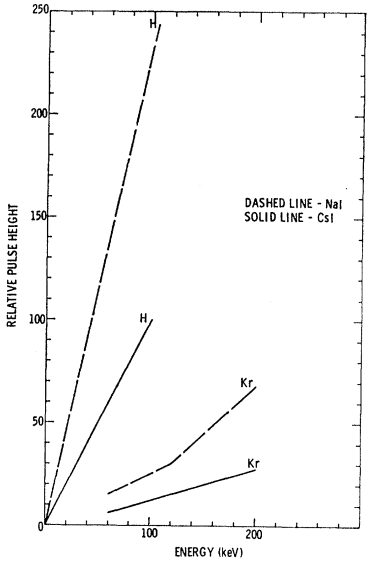

FIG. 7. Least-squares curves of relative pulse height from NaI(Tl) and CsI(Tl) versus particle energy for H and Kr for comparison.

In 1953 Allison and Casson[5] investigated the scintillations from NaI(Tl) using monoenergetic H, D, He, and Ne in the energy range 60–600 keV. By using the PH of 100 keV H^+ to normalize their data to ours we find excellent agreement with the present He data where the energies overlap. Our Ne line and the Allison-Casson Ne line overlap almost exactly from 150–200 keV. The present Ne line has a lower slope, thus a slope change appears around 180 keV. However, considerable uncertainty can arise in normalizing one set of data with respect to another so that whether or not a break occurs is questionable. Similarly our CsI(Tl) results[1] not only overlap those of Bashkin *et al.*,[7] but also extend significantly further. Hill[6] did similar work with H, H_2, and He in the energy range 4–60 keV. Hill presents a nonlinear display of PH versus E for He in this energy range, but two intersecting straight lines can be drawn equally well through his NaI(Tl) data.

A comparison of the energy resolution by CsI(Tl)[1] with the energy resolution by NaI(Tl) as displayed in Fig. 5 shows a number of important differences. For H^+, the resolution by NaI(Tl) is 17% at 100 keV and 100% at 20 keV. This is better by 5% at the high energy end and by 30% at the low energy end than by CsI(Tl). For D^+, the resolution is almost the same for the two crystals. In contrast, the resolution by NaI(Tl) for ions of mass greater than two amu is worse than by CsI(Tl), and the difference increases rapidly as ion mass is increased. It is believed that the increase in pulse width of NaI(Tl) is due to the presence of what appears to be two pulses in one. Figure 8 shows a pulse-height distribution from NaI(Tl) bombarded with 60 keV C^+ ions. It is seen that the distribution, which is typical for ions not fully stripped of electrons, is asymmetric and is perhaps the composite of two pulse-height distributions in coincidence. It is not known why two arise. It may be that one is due directly to the Na and the other to the I as both of these release the energy, obtained from the incident ion, through the Tl impurity. This supports the observation that the break in slope of PH versus E is related to the Na-I mass difference. Cs and I have essentially the same mass so that the slope changes and pulse coincidences either are not present or are not noticeable. Other crystals whose two main constituents are of very different mass should be investigated.

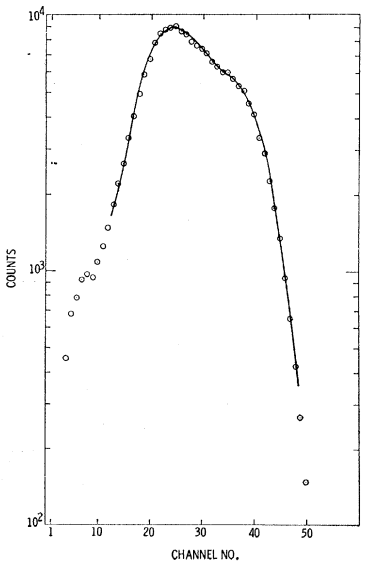

FIG. 8. Pulse height distribution from NaI(Tl) bombarded with 60 keV C^+ ions. Note the appearance of two pulse height distributions in coincidence.

V. INTERPRETATION

Bohr,[11] in an extended treatment of the penetration of atomic particles through matter, has examined the problem of energy loss by atoms of small initial velocity in a stopping medium. Atomic particles approaching each other with relative velocities less than the characteristic Bohr velocity, $V_0=e^2/h$ where e is the electron charge unit, will experience a quasiadiabatic collision. The main energy transfer occurs elastically with the resident atom of the medium recoiling. Excitation and ionization also occur, but these inelastic energy losses are minor in comparison to the elastic losses. His classical treatment is appropriate to this experiment as the velocities of the ions used were in the range $0.1V_0 \lesssim V \lesssim 2V_0$.

In a recent paper,[15] range-energy equations were developed for heavy ions incident on a monatomic gas, which utilize Bohr's theory on atom-atom elastic collisions and the work of Lindhard, Scharff, and Schiott[16] on atom-electron inelastic collisions. Those equations apply here equally well. From the expression for stopping power

$$dE/dx=N(S_n+S_e), \tag{1}$$

where N is the number of atoms per cm^3 in the stopping medium, S_n and S_e are the atomic and electronic stopping cross sections, respectively, the stopping power of the target material for the incident ion and also the range of the ion in the material can be obtained.

The Bohr atomic stopping cross section is given simply by

$$S_n(E_1)=\tfrac{1}{2}\sigma E_m, \tag{2}$$

where σ is the collision total cross section given by Bohr as an approximation for shielded Coulomb fields and E_m, E_1 are the maximum energy transferred in a direct collision and the energy of the incident particle, respectively. The Lindhard, Scharff, and Schiott[16] electronic stopping cross section is closely approximated by

$$S_e=8\pi a[Z_1^{7/6}Z_2e^2/(Z_1^{2/3}+Z_2^{2/3})](V/V_0), \tag{3}$$

where V is the velocity of the incident particle. Subscripts 1 and 2 refer to the incident and target particles, respectively.

[15] G. L. Cano and R. W. Dressel, Phys. Rev. **139**, A1883 (1965).
[16] J. Lindhard, M. Scharff, and H. Schiott, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **33**, No. 14, 1 (1963).

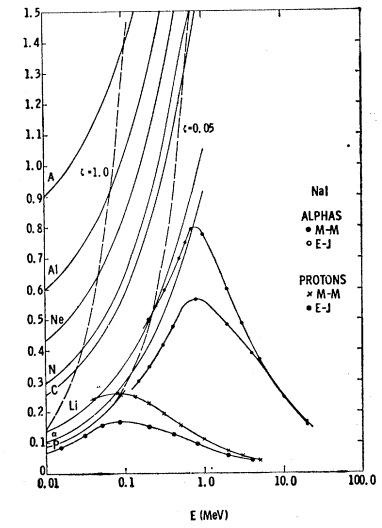

FIG. 9. Calculated stopping power of NaI versus incident particle initial energy. The Meyer-Murray and Eby-Jentschke curves for alpha particles and protons are shown for comparison.

The stopping power as given by Eqs. (1), (2), and (3) is, to a close approximation,

$$\frac{dE}{dx}=K_1\left[\frac{Z_1Z_2M_1}{(Z_1^{2/3}+Z_2^{2/3})^{1/2}(M_1+M_2)}\right]+K_2\left[\frac{Z_1^{7/6}Z_2}{(Z_1^{2/3}+Z_2^{2/3})^{3/2}}\right]V\frac{\text{keV}}{\text{g/cm}^2},\quad(4)$$

where

$$K_1=1.25\times10^9\pi a_0e^2N/\rho\epsilon$$

and

$$K_2=5\times10^9\pi a_0e^2N/(\rho V_0).$$

ϵ is the base of the natural logarithms, ρ is the stopping material density, and cgs units are used unless otherwise indicated. It should be noted that Eq. (4) is valid only if $\zeta\gtrsim1$ and $\kappa\gg\zeta$ where ζ is the ratio of the collision diameter to screening parameter for the two interacting particles, and κ is the ratio of collision diameter to rationalized deBroglie wavelength for the reduced mass particle.

A projectile particle incident on a crystal loses energy inelastically in knocking resident ions from their lattice site. The cumulative energy loss by the incident particle due to the binding energy of 7.1 eV for NaI and 6.0 eV for CsI is at most 5% of the initial energy value. The basis of this assertion is a range calculation made from Eq. (4) for the incident ions most penetrating into the crystal and then estimating the cumulative inelastic energy loss by the incident particle in releasing an ion per atomic layer passed from its lattice site. For 100-keV C^+ ions into either crystal the loss, approximately 0.02 MeV/mg cm^{-2}, is at most 5 keV in NaI and 4 keV in CsI. This type of energy loss is not included in the derivation of Eq. (4). Figure 9 shows plots of dE/dx as obtained from Eq. (4), plus 5% as a binding-energy correction, for a number of ions incident on NaI. The dashed lines are for $\zeta=1$ and $\zeta=0.05$. In the region $\zeta>1$, the treatment is valid, but for $0.05\leq\zeta<1$ it may be in error, particularly close to $\zeta=0.05$ for low-mass particles. Included in this figure are the plots of dE/dx versus E for alpha particles and protons in NaI of Eby and Jentschke[8] and Murray and Meyer.[9] It is seen that the E-J curve agrees within 5% with the M-M curve for $E\geq5$ MeV. Below 5 MeV, however, the difference is as much as 50%. The values obtained from Eq. (4) in its region of applicability for alpha particles and protons fall between the E-J and M-M curves. The agreement is within 15% with either curve. Figure 10 is of plots similar to Fig. 9, but for CsI. Here the agreement is not as close.

The range-velocity relation as obtained from Eq. (4) is

$$R=\frac{2E_0}{kN}\left[\frac{V}{V_0}-\frac{K}{2\epsilon k}\ln\left(1+\frac{2\epsilon k}{K}\frac{V}{V_0}\right)\right],\quad(5)$$

where

$$E_0=\tfrac{1}{2}M_1V_0^2,$$
$$k=8\pi ae^2Z_1^{7/6}Z_2(Z_1^{2/3}+Z_2^{2/3})^{-1},$$
$$K=4\pi ae^2Z_1Z_2M_1(M_1+M_2)^{-1}.$$

One can now calculate the contributions to the total energy loss by the incident particle in elastic atom-atom and in inelastic atom-electron interactions by modifying Eq. (1). To compute the amount of energy loss in the inelastic processes omit S_n in Eq. (1), integrate, and use R, the total range of the incident particle in the crystal, from Eq. (5). This gives a close approximation to the total energy of the incident particle going into inelastic light-producing processes in the crystal. By following this procedure and assigning to the inelastic energy losses the pulse heights of Figs. 2 and 6, Figs. 11 and 12 were obtained.

Figure 12 for CsI, shows that two universal curves are obtained; namely, one for the ions completely

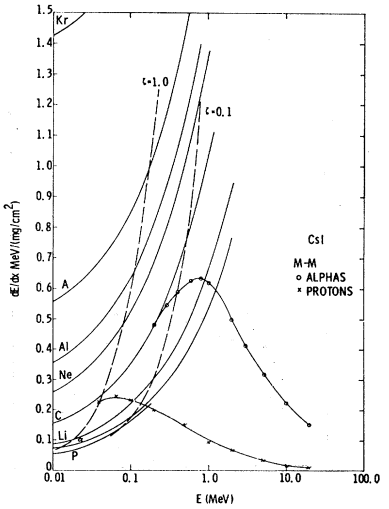

FIG. 10. Calculated stopping power of CsI versus incident particle initial energy. The Meyer-Murray curve for alpha particles and protons is shown for comparison.

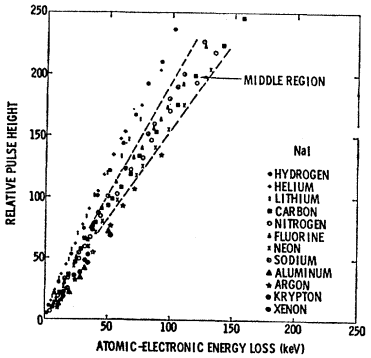

FIG. 11. Pulse height from NaI(Tl) versus calculated particle-energy-loss in atom-electron inelastic collisions. Three regions are defined.

stripped of electrons and one for the ions incompletely stripped of electrons. This figure indicates clearly that if a fixed amount of energy is expended by an atomic particle of mass ≥ 12 amu in CsI in atom-electron interactions, the PH from the crystal is only slightly different from the PH produced by any atomic particle of mass ≥ 12 amu that expends an equal amount of energy in atom-electron processes in the crystal. Also, it indicates that for a given initial energy the greater the incident particle mass, the greater the amount of energy loss in negligible light-producing interactions. The H, D, and He PH versus E relations fall on a line different from that of the higher mass ions. Possible reasons for this follow.

The efficiency for elastic energy transfer from the incident ions of mass ≥ 12 amu to the CsI ions is several orders of magnitude greater than that from H^+ and D^+. This is determined on the basis of the physical cross-sectional area of the incident particle and its energy transfer to a Cs or I ion in a direct encounter. Because H^+ and D^+ are bare nuclei, have low mass, and velocity

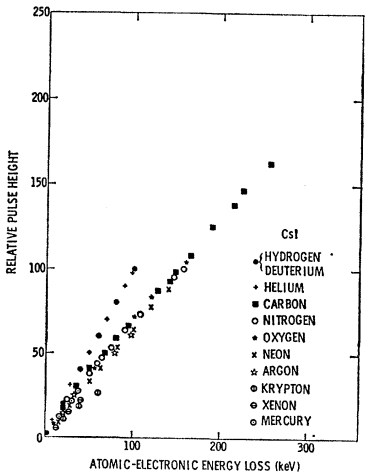

FIG. 12. Pulse height from CsI(Tl) versus calculated particle-energy-loss in atom-electron inelastic collisions. Two universal lines are defined.

close to V_0, they lose their energy predominantly by coulombic nucleus-electron and not nucleus-atom encounters. Two explanations offered as to why the He line falls on the H^+, D^+ line are as follows: (1) Possibly along a large part of its path in the crystal, the He is stripped of electrons; (2) because the efficiency for elastic energy-transfer by the He^+ to the CsI ions is a factor of about 10 less than by the heavier ions and as the He^+ line at 100 keV (Fig. 6) is only about 5 keV from the H, D line, this efficiency difference is enough to produce coincidence of the lines.

Figure 11, for NaI, shows three regions into which the pulse heights fall. The left one includes H, D, and He and Li; the middle one includes the ions of mass 12 amu$\leq M_1 \leq$23 amu and the right one includes ions of mass 27 amu$\leq M_1 \leq$133 amu. The same hypotheses are offered to explain why the particles of mass 1–7 amu define the left region that were given in the case of CsI. For the particles in the middle group it may be that they transfer their energy preferentially to the Na ions while the particles of mass equal to or greater than 27 amu do this for some unknown reason to the I ions. Thus, two additional lines arise. On this basis only one line appears in the CsI case for 12 amu$\leq M_1 \leq$202 amu because the masses of the crystal ions are essentially the same.

In conclusion, for the low-energy, high-mass region of this experiment the saturation mechanism and the theory which couples the nuclear and electronic collisions presented here may be complementary. Saturation still may occur to some extent in our case; however, it does not appear to be the dominant effect. It is seen in Figs. 9 and 10 that for a given crystal the stopping power, dE/dx, reaches a maximum value at an energy characteristic of the incident ion. The saturation model applies at energies which correspond to the high-energy side of the peak of the dE/dx curves. At these high energies the incident ion transfers its energy mostly through inelastic interactions which may ultimately saturate the impurity, light-producing centers. On the low-energy side of the peak the energy is transferred by the incident ion to the crystal predominantly through elastic collisions in which light is produced with negligible efficiency. As an example, the greatest curvature in the PH versus E curve for alpha particles on NaI(Tl) occurs at about 3 MeV.[8] This is on the high-energy side of the corresponding stopping power peak, Fig. 9. A crystal does not begin to saturate at a fixed value of dE/dx. This depends on the particle and the particle energy.

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