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Scintillation response of nonactivated and activated NaI to random and channeled ions: Influence of the luminescence mechanism*

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Previous experiments have shown that the scintillation response to random positive-ion bombardment of alkali iodides, both nonactivated and activated, have similar characteristics even though the luminescence mechanisms are very different. This paper reports experiments on nonactivated NaI and NaI(Tl). The relative pulse height as a function of energy for randomly incident protons (3–14 MeV) and ^{16}O ions (5–50 MeV) for both crystals has been recorded. Also, pulse-height spectra due to energetic ^{16}O ions (10–50 MeV) channeled along the {100} plane and ⟨100⟩ axis of nonactivated NaI at 100°K are presented and compared with those obtained previously for NaI(Tl). The following observations are made: (i) The ratio of the relative number of photons per unit energy for ^{16}O ions to that for protons is very nearly the same for both crystals. (ii) The ratio of light outputs for totally channeled particles to random particles is nearly the same for both crystals. (iii) The detailed characteristics of pulse-height spectra from channeled ions are very similar for the two crystals. These results indicate that the scintillation response is quite insensitive to the particular luminescence mechanism.

I. INTRODUCTION

The response of alkali-iodide scintillators to energetic ion bombardment has been the subject of numerous investigations. Newman *et al.*[1,2] studied in detail the scintillation pulse-height characteristics of NaI(Tl) and CsI(Tl) to energetic heavy ions. They demonstrated that scintillation efficiency decreases with increasing specific energy loss of the incident ion, and that it also depends on the ion identity. Blue and Liu[3] showed that the relative pulse-height response for all alkali-iodide crystals, both nonactivated (at 77 °K) and activated (at 300 and 77 °K), is nearly linear for MeV protons and is nonlinear for MeV α particles. (Nonactivated alkali halides scintillate only at low temperatures.) The observed similarity in the nonactivated and activated crystal response is particularly interesting, since the luminescence mechanisms in the two cases are very different.

A scintillation pulse is initiated by ionizing radiation that produces electrons, holes, and excitons. In nonactivated NaI, light emission is due to radiative decay of the self-trapped exciton,[4,5] an excited state of the crystal formed either by recombination of an electron with a self-trapped hole or by relaxation of a "free" exciton to the self-trapped state (having point-group symmetry D_{2h}). The radiative transition probability of the self-trapped exciton is appreciable only at low temperatures, and in NaI it has a maximum value near 70 °K.[5] The emission spectrum is composed of a single band with a peak at 295 nm. It should be emphasized that this emission is *intrinsic* and arises from an excited state of the pure crystal; it is not associated with a vacancy, interstitial, or other defect. On the other hand, luminescence from activated alkali-halide crystals results from the radiative decay of an excited state of the activator ion. In NaI(Tl) the emission spectrum observed upon excitation by ionizing radiation is a single band with a peak at 420 nm, and arises from an excited state of the Tl^+ ion. Recent experiments[6] utilizing time-resolved emission and absorption spectroscopy have shown that the Tl^+ emission is stimulated by sequential capture of an electron and hole (in either order) at a Tl^+ site. The radiative transition probability is relatively high at room temperature, and NaI(Tl) scintillators are commonly used at room temperature. Thus, in NaI(Tl), luminescence arises from electron-hole recombination at the impurity site and is an *extrinsic* process.

An account of the observed behavior of activated alkali iodides subject to heavy-ion bombardment was provided by Meyer and Murray[7] and extended by Luntz and Heymsfield.[8] These analyses employ ion track-effect models which are independent of the details of the luminescence process.

Altman *et al.*[9] extended the study of NaI(Tl) scintillation response to include channeled ions. They found that ions which penetrate along a major symmetry direction produce more scintillation photons than those which undergo random trajectories. This was a direct verification of the predictions of Luntz and Bartram[10] who argued that the reduction in stopping power due to channeling should be accompanied by an enhancement of light production efficiency.

The objective of the present work is to investigate the influence of the luminescence mechanisms on the scintillation response of nonactivated NaI

and NaI(Tl), especially with respect to channeled ions. These crystals are well suited to such a study, as the luminescence mechanisms are well understood, the scintillation efficiencies are high, and the response of NaI(Tl) to channeled ions is established and exhibits sufficient complexity to provide the means for detailed comparison. To this end, the results of relative pulse-height measurements for nonactivated NaI and NaI(Tl) are presented as a function of energy for randomly incident protons and ^{16}O ions. Also, pulse-height spectra due to energetic ^{16}O ions (10–50 MeV) aligned along the {100} plane and ⟨100⟩ axis of nonactivated NaI at 100 °K are presented and compared with those obtained in previous experiments on NaI(Tl).[9]

II. EXPERIMENT

The experiments reported here were performed with the Ballistic Research Laboratories tandem Van de Graaff accelerator at Aberdeen Proving Ground. The experimental setup and crystal-mounting technique are described in the preceding paper.[11] There were, however, two differences in the experimental arrangement that should be noted. Since nonactivated NaI is an efficient scintillator only at low temperature, the goniometer was modified to incorporate a cold finger which permitted the crystal to be cooled to 100 °K. Owing to the cooling, it was not possible to couple the crystal directly to the face of the photomultiplier tube. Consequently, the resolution of the detection system was not as good as previously obtained[11] and thus no triple-peaked spectra were observed.

All NaI(Tl) and nonactivated crystals were obtained from Harshaw. The temperature of the nonactivated crystal during all experiments was measured to be 100 ± 2 °K by a thermocouple.[12] All measurements on NaI(Tl) were performed with the crystal at room temperature, but using the same goniometer mount as for nonactivated NaI. Consequently, the resolution for NaI(Tl) data was approximately the same as for nonactivated NaI. When measuring the random peak pulse height as a function of energy, an auxiliary encapsulated NaI(Tl) crystal containing ^{241}Am was mounted on the face of the photomultiplier tube to serve as a pulse-height standard. Thus all spectra contained a peak due to ^{241}Am α counts. By measuring the peak pulse-height positions due to protons and ^{16}O ions relative to the ^{241}Am α peak, possible errors due to gain shifts in the photomultiplier and electronics were eliminated.

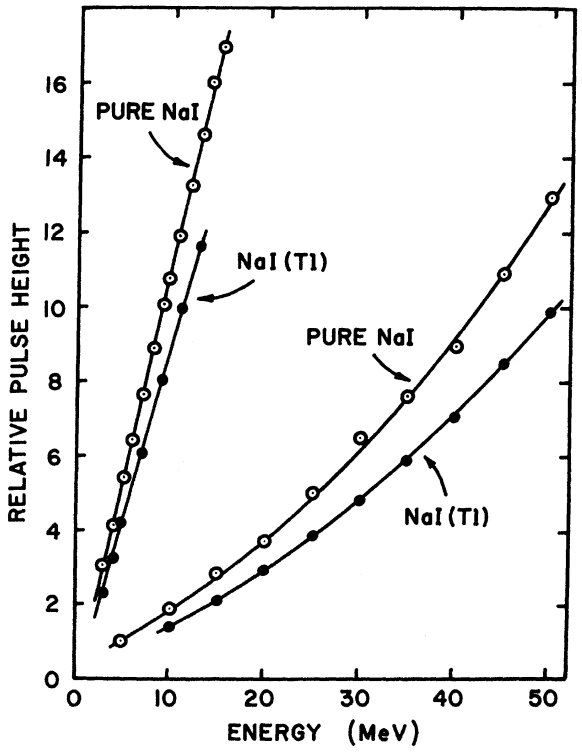

FIG. 1. Peak pulse height versus energy for protons and ^{16}O ions for random incidence on nonactivated NaI at 100 °K and NaI(Tl) at room temperature.

III. RESULTS AND DISCUSSION

A. Random pulse height as a function of energy: Nonactivated and activated NaI

The peak pulse height as a function of energy was measured for protons in the 3–14-MeV range and for ^{16}O ions in the 5–50-MeV range for random incidence on both nonactivated NaI and NaI(Tl). The results of experiment are shown in Fig. 1. The proton response is essentially linear for both crystals with the nonactivated crystal producing a larger pulse than NaI(Tl) for equal energies. The response to ^{16}O ions is clearly nonlinear with the points curving away from the energy axis for both crystals. Here, again, larger pulses are produced in nonactivated NaI than in NaI(Tl). The shapes of the response functions are the same for both crystals indicating that the scintillation response characteristics are independent of the particular luminescence mechanism and depend only on the identity and energy of the incident ion.

It should be noted that the relative number of photons per unit energy, L/E, for ^{16}O ions is substantially less than that for protons, for both nonactivated NaI and NaI(Tl). However, the ratios of L/E for ^{16}O to that for protons at 10 MeV are nearly identical, 0.17 for nonactivated NaI and 0.16 for NaI(Tl), respectively. Thus the decrease in L/E is not to be identified with a saturation of activator

centers, a conclusion consistent with that drawn by Gwin and Murray[13] on the basis of studies in cesium-iodide crystals. We conclude that the decrease in L/E is a characteristic of the charged-particle interaction with the target.

B. Random and Channeled pulse-height spectra: Nonactivated NaI

Figure 2 shows scintillation pulse-height spectra due to 20-MeV ^{16}O ions incident on nonactivated NaI in a random direction and with the beam aligned along the {100} plane and ⟨100⟩ axis. The random spectrum has a characteristic Gaussian shape due to the instrumental resolution of the photomultiplier detection system. Both aligned spectra have a peak at approximately the same pulse height as the random spectrum and in addition a second peak (the channel peak) at a pulse height approximately 40% greater than that of the random peak. The channel peak is attributed to particles which remain channeled for their entire trajectory. There is also a continuous distribution of pulses between the two peaks, attributed to particles which are initially channeled and subsequently dechanneled at some point along their trajectory. The channel peak for the axial spectrum is significantly larger than for the planar spectrum indicating that a larger fraction of particles is channeled to the end of their range. However, it should be noted that the pulse height corresponding to the channel peak is approximately the same for both aligned spectra. This indicates that the pulse height due to ions that remain channeled for their entire trajectory along the ⟨100⟩ axis and {100} plane is approximately the same.

In Fig. 3, scintillation pulse-height spectra for

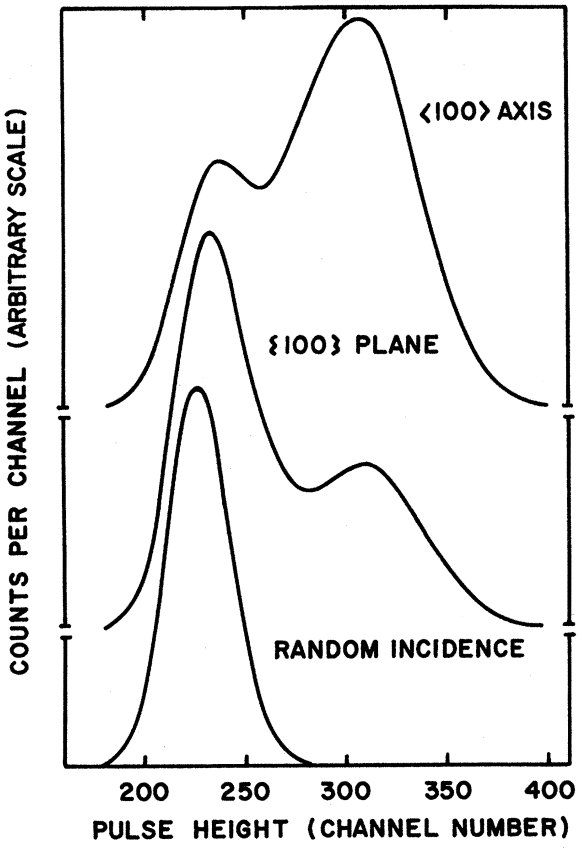

FIG. 2. Pulse-height spectra from 20-MeV ^{16}O ions on nonactivated NaI for random incidence and for the beam aligned along the {100} plane and ⟨100⟩ axis with the crystal at 100 °K.

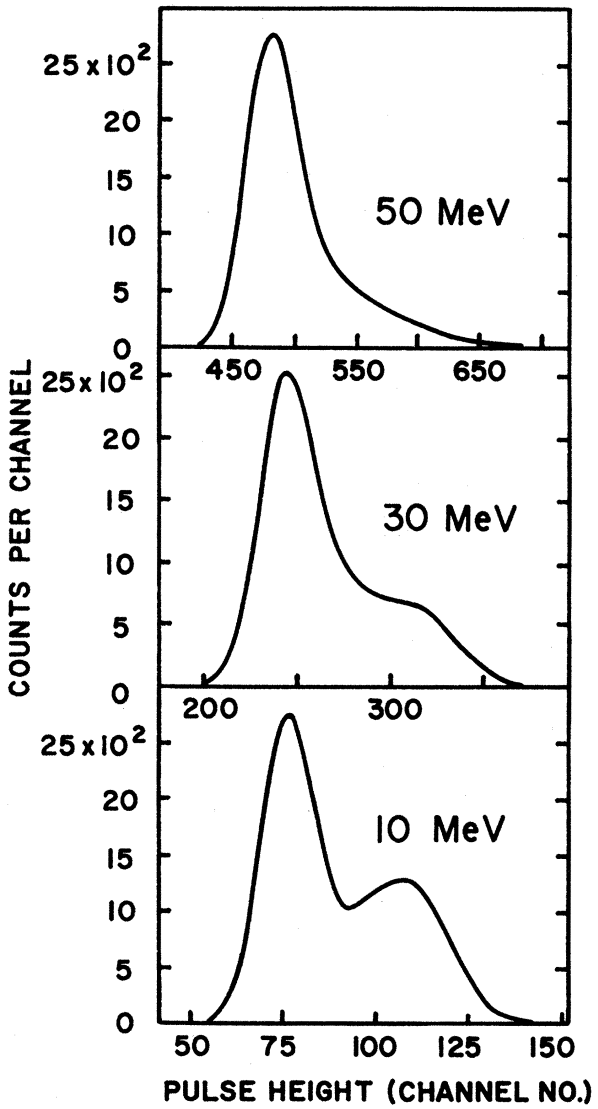

FIG. 3. Pulse-height spectra from 10-, 30-, and 50-MeV ^{16}O ions on nonactivated NaI along the {100} plane with the crystal at 100 °K.

^{16}O ions aligned along the {100} plane for incident energies of 10, 30, and 50 MeV are shown. As the incident energy increases, the number of pulses in the channel peak is clearly reduced and there is no well-defined channel peak for the 50-MeV spectrum. In terms of the parameter R defined in the preceding paper,[11] increasing E results in an increase in R and a corresponding decrease in channeling stability. Therefore, the relative number of counts in the channel peak is expected to decrease with increasing energy, as observed. It should be noted that R was derived for axial channeling. However, it is easily shown that the energy dependence of R is the same for planar channeling.[12]

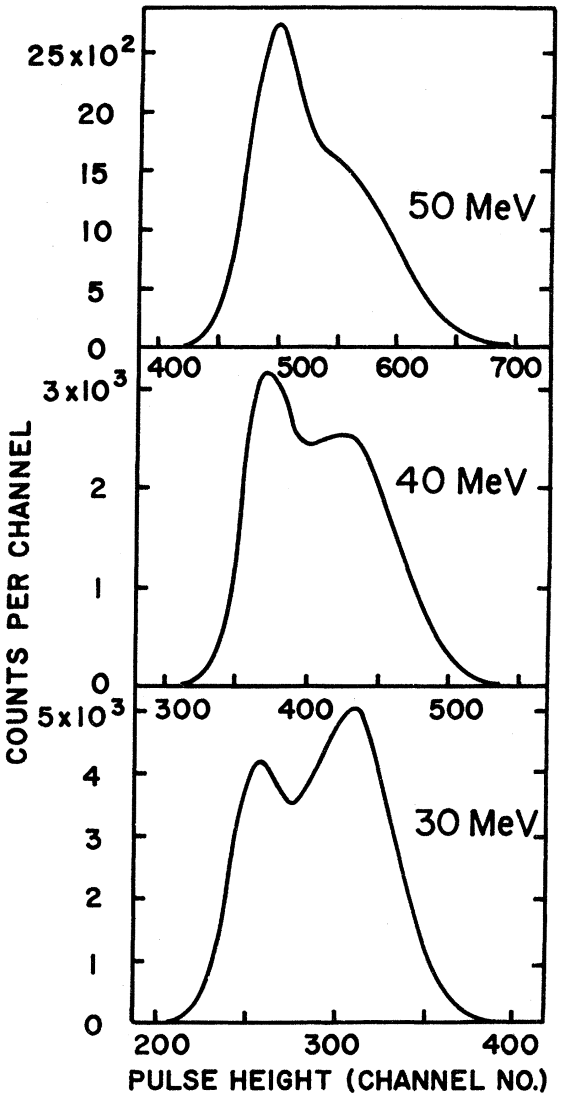

FIG. 4. Pulse-height spectra from 30-, 40-, and 50-MeV ^{16}O ions on nonactivated NaI along the ⟨100⟩ axis with the crystal at 100 °K.

Scintillation pulse-height spectra for ^{16}O ions aligned along the ⟨100⟩ axis for incident energies of 30, 40, and 50 MeV are shown in Fig. 4. As in the planar case, the relative number of counts in the channel peak decreases with increasing energy. However, the channel peak is larger than the random peak for all spectra up to 30 MeV. At 40 MeV the random peak becomes dominant, and at 50 MeV there is no clearly defined channel peak. Figures 3 and 4 demonstrate that, in general, the fraction of particles channeled to the end of their range is greater for ⟨100⟩ axial channeling than for {100} planar channeling.

C. Comparison of nonactivated NaI and NaI(Tl) channeling spectra

Scintillation pulse-height spectra for nonactivated NaI at 100 °K may be compared with spectra obtained by Altman *et al.*[9] for NaI(Tl) at room temperature. In Fig. 5, the scintillation pulse-height spectra for 10-MeV ^{16}O ions aligned along the {100} plane for both nonactivated NaI and NaI(Tl) are

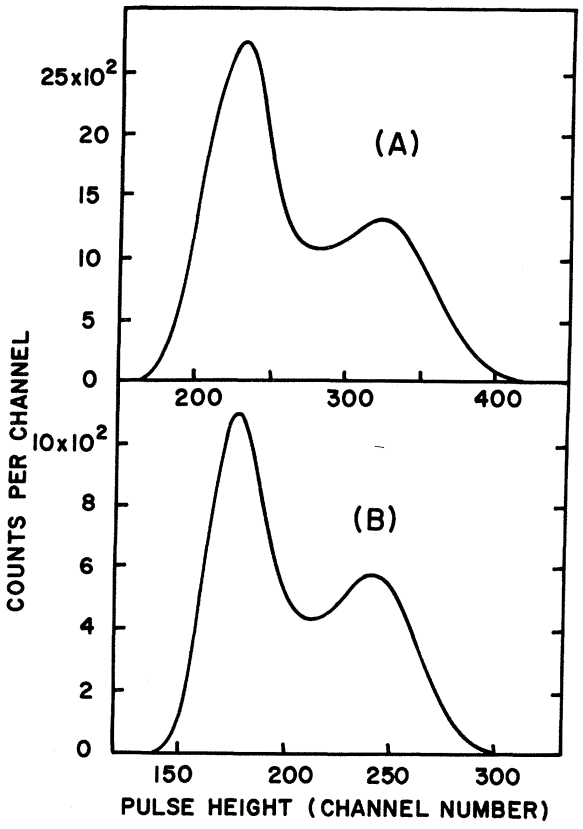

FIG. 5. Pulse-height spectrum from 10-MeV ^{16}O ions along the {100} plane; (A) nonactivated NaI with crystal at 100 °K (B) NaI(Tl) with the crystal at room temperature [from Altman *et al.* (Ref. 9)]. The abscissas of (A) and (B) can be directly compared.

shown. The important feature to note is that both spectra have similar shapes, indicating that the general character of the scintillation response is the same. Further, the ratio of the average pulse height of particles channeled for their entire trajectory L_c, obtained in the manner of Altman *et al.*,[9] to the random pulse height L_R, is approximately 1.4 for both crystals. This clearly indicates that the luminescence mechanism has little influence on the ratio L_c/L_R.

Spectra in the 10–50-MeV range for ^{16}O ions aligned along the {100} plane and ⟨100⟩ axis have been compared and the similarity in spectral shape is the same as in Fig. 5. This demonstrates that the effect of channeling on the scintillation pulse-height response to energetic heavy-ion bombardment of both nonactivated and activated NaI is insensitive to the particular luminescence mechanism and appears to be associated primarily with the reduction in stopping power.

IV. CONCLUSIONS

The experiments reported here show that the scintillation response to positive ions of both NaI(Tl) and nonactivated NaI are very nearly the same in the following respects:

(i) The ratio of the relative number of photons per unit energy, L/E, for ^{16}O ions to that for protons is very nearly the same for both crystals and is substantially less than unity.

(ii) The ratio of light output for totally channeled particles to random particles, L_c/L_R, is nearly the same for both crystals.

(iii) The detailed characteristics of pulse-height spectra from channeled ions are very similar for the two crystals for channeling along the ⟨100⟩ axis and {100} plane.

These results indicate that the scintillation response to positive ions is quite insensitive to the particular luminescence mechanism, i.e., whether intrinsic self-trapped exciton luminescence as in nonactivated NaI, or impurity luminescence as in NaI(Tl). In light of this and previous work,[3] it is reasonable to attribute the behavior of the scintillation response to positive ions to a single common process. Luntz[8] has suggested that the region close to the track of a highly ionizing particle penetrating through a phosphor is rendered relatively ineffective as a scintillating medium owing to radiation-damage effects, and that the scintillation response is dominated by the fraction of energy deposition outside the inner track. Thus, the proposed common feature affecting the response to positive-ion bombardment is the enhancement of damage-related competitive processes by high ionization density close to the ion track. The exact nature of the damage process and how it reduces scintillation efficiency is not known. We do know, however, from a number of studies,[14,15] that a variety of point defects with lifetimes in the range of 10^{-9} to about 10^{-3} sec are created by ionizing radiation. Such defects could inhibit luminescence by (a) serving as transient traps that capture electrons, holes or excitons, (b) reduction of radiative decay probabilities of luminescence centers, and (c) absorption of emitted luminescence photons. The creation of such transient defects is expected to occur in both activated and nonactivated crystals and would, therefore, influence the scintillation behavior in a similar way.

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