Scattering of Slow Neutrons from Propane Gas*

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(Received September 18, 1961)

Measurements of the partial differential neutron scattering cross sections for room-temperature propane gas are reported. These measurements were made at incident energies of 0.0101, 0.0254, 0.0736, and 0.102 ev at seven scattering angles between 16.3° and 84.7° using the Materials Testing Reactor phased chopper velocity selector. The data are converted to the scattering-law presentation and compared with three theoretical calculations: (a) The ideal gas, using an effective mass obtained from an average of the mass tensors for the three types of H atoms in propane, gives poor agreement. (b) The Krieger-Nelkin approximation, which includes the effect of zero-point vibrations, gives limited agreement for energy transfer less than $0.5k_BT$ at intermediate momentum transfers. At large momentum transfers where vibrational effects become important it underestimates the cross section. (c) A modification of the Krieger-Nelkin theory that includes the effects of single-quantum transitions from the three lowest vibrational states gives better agreement. The discrepancies still present at large momentum and energy transfers are attributed to an uncertainty in the methyl-group barrier height for the three lowest energy modes, to the harmonic oscillator approximation for these modes, and to the approximate molecular orientation averaging used in the calculation.

I. INTRODUCTION

PARTIAL differential cross sections have been measured for scattering of slow neutrons from gaseous propane at room temperature and at relatively low pressures. The incident neutron energies used were 0.0101, 0.0254, 0.0736, and 0.102 ev.

Previous neutron studies of propane have yielded only total cross sections. Carroll[1] measured the total neutron cross section of propane with cadmium-filtered neutrons, while Melkonian[2] obtained energy-dependent total cross sections from 0.003 to 10 ev.

The present measurements are similar to those of Randolph et al.[3] for methane in which the effects on the scattering of the vibrational and rotational states were observed. Because of the translational motion in a gas, these effects are not observed as sharp peaks in the data. In propane the rotational states are much more closely spaced than in methane and effects due to their discrete character should appear only at the lowest energies and most forward angles. With these incident energies, vibrational transitions induced by neutron collisions played no role in methane, but such processes involving the lowest vibrational states are expected to be important in propane. That this is true is evident in the data when it is compared to theory.

II. EXPERIMENT, APPARATUS, AND SAMPLES

To perform the experiment, bursts of monoenergetic neutrons were obtained from a Materials Testing Reactor beam using the MTR slow-neutron velocity selector.[4] These neutrons are scattered by the sample and detected in BF_3 counters two meters from the sample at seven scattering angles from 16° to 84° with respect to the incident beam. The pulses from the counters are

time-of-flight analyzed and stored in a 4096-channel analyzer. Ten-microsecond channel widths were used.

The experimental procedure and method of data analysis as well as the type of sample containers used were basically the same as employed in the methane experiment.[3] The samples consisted of research grade propane with a mole percent purity of 99.9+. The most probable impurity is ethane which can account for less than 0.1% of the total scattering. The sample filling pressure was adjusted to give approximately a 90% transmission for each experimental run with the exception of the 0.0101-ev run. In the latter case the sample pressure was adjusted to give a transmission of 75% in order to obtain a higher counting rate. No effects that are attributed to multiple scattering are evident in the data due to this thicker sample. For the data runs of 0.0101, 0.0254, 0.0736, and 0.102 ev, the filling pressures used were 45, 15, 30, and 50 psig, respectively. At these pressures the molecular mean
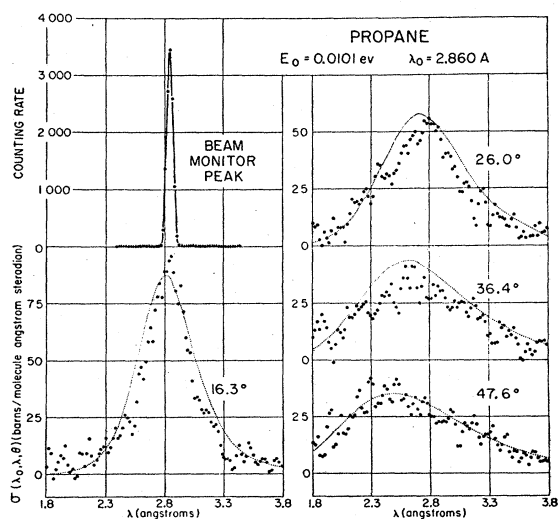

FIG. 1. Partial differential cross sections for scattering 0.0101-ev neutrons from room temperature propane. The dotted curve is the modified Krieger-Nelkin theory. The theoretical curves have not been resolution broadened.

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[1] H. Carroll, Phys. Rev. **60**, 702 (1941).
[2] E. Melkonian, Phys. Rev. **76**, 1750 (1949).
[3] P. D. Randolph, R. M. Brugger, K. A. Strong, and R. E. Schmunk, Phys. Rev. **124**, 460 (1961).
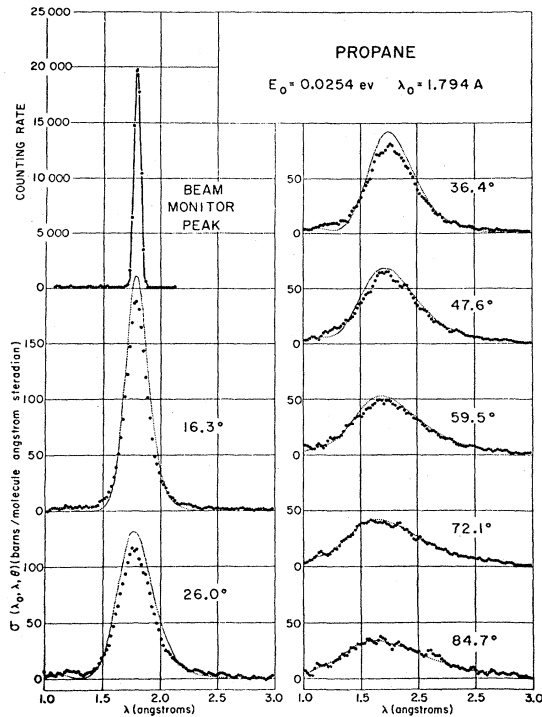[4] R. M. Brugger and J. E. Evans, Nuclear Instr. and Methods **12**, 75 (1961).

FIG. 2. Partial differential cross sections for scattering 0.0254-ev neutrons from room temperature propane. The dotted curve is the modified Krieger-Nelkin theory. The theoretical curves have not been resolution broadened.
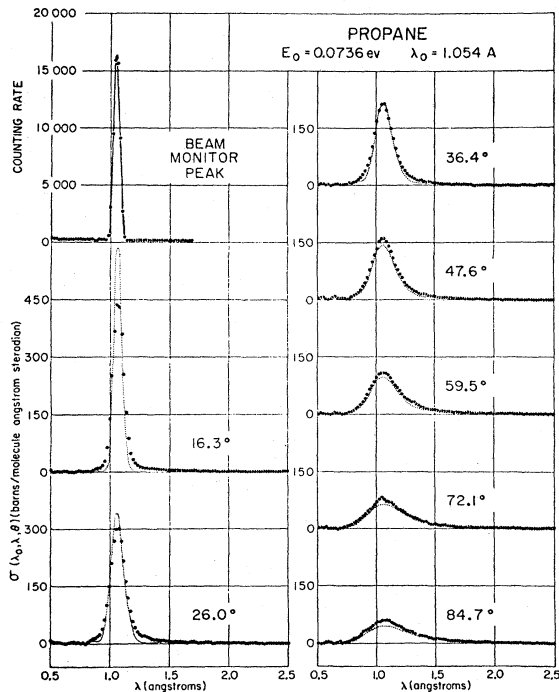
free path is still large so that changes in pressure are not expected to affect the data. The wall thickness of the containers in the area of the beam was 0.010 in. in all cases.

III. RESULTS

Figures 1 through 4 show the cross sections per propane molecule differential in wavelength and solid angle vs wavelength for 0.0101, 0.0254, 0.0736, and 0.102 ev incident neutron energies. The dotted curves are based on a Krieger-Nelkin[5] theory modified so that effects due to single vibrational transitions are included. In Fig. 1, the reasons for the slight differences in the wavelength positions of the peaks of the theoretical curves and the experimental curves are not known.

The incident energies, though pre-set by the chopper phases, are determined from the position in time of the beam monitor peak. In this experiment the 0.0736-ev data and 0.0254-ev data were taken with a BF_3 counter in the forward beam as a monitor. It was found that this type counter was subject to burnout in the high gamma field of the beam. A fission chamber replaced the BF_3 counter and was used for the 0.0101- and 0.102-ev data. After this change, the absolute cross-section calibration of the instrument was accomplished by running a methane sample at 0.07 ev and comparing with previous scattering law results.[3]

FIG. 3. Partial differential cross sections for scattering 0.0736-ev neutrons from room temperature propane. The dotted curve is the modified Krieger-Nelkin theory. The theoretical curves have not been resolution broadened.
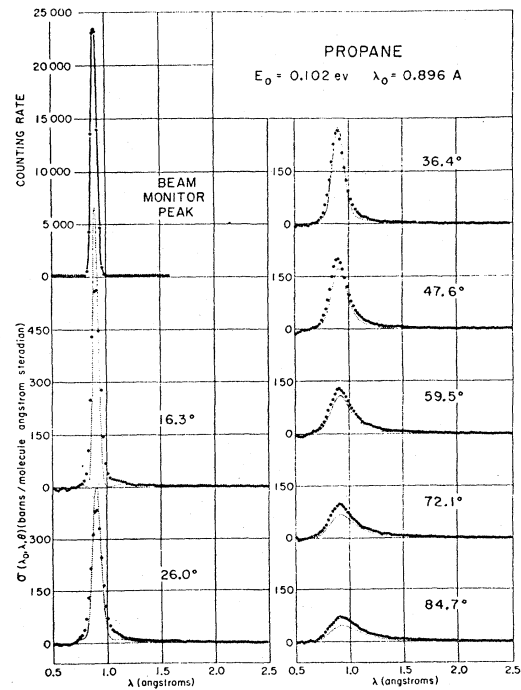

FIG. 4. Partial differential cross sections for scattering 0.102-ev neutrons from room temperature propane. The dotted curve is the modified Krieger-Nelkin theory. The theoretical curves have not been resolution broadened.

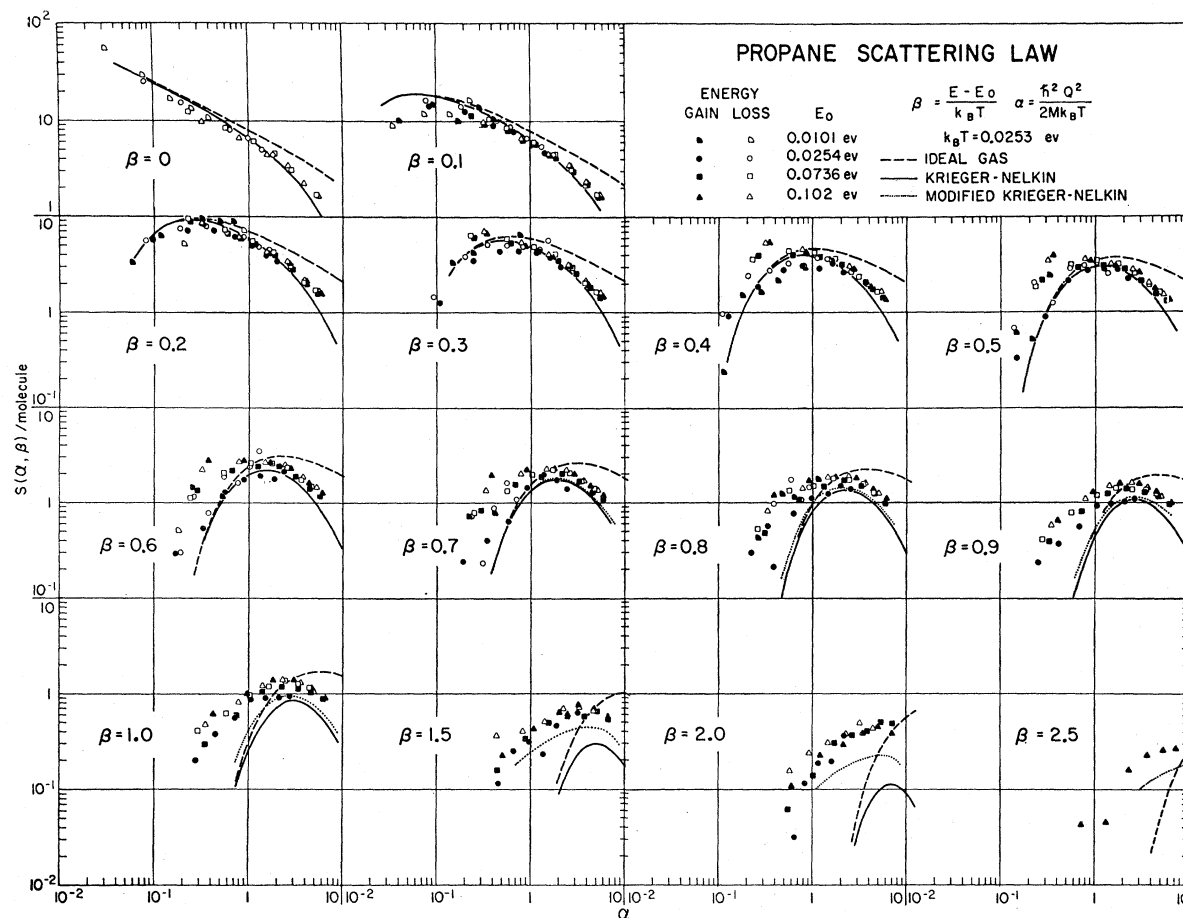[5] T. J. Krieger and M. S. Nelkin, Phys. Rev. 106, 290 (1957).

FIG. 5. Propane scattering law. The effective mass used to convert the partial differential cross sections to $S(\alpha,\beta)$ is $M = 1$ hydrogen atomic mass.

The counting statistics for the 0.0736- and 0.102-ev data are better than those at the lower energies because the initial neutron energies are nearer the peak of the reactor thermal flux. The fluctuation of the points gives an indication of the counting statistics after "5 point smoothing"[3] of the raw data.

Figure 5 shows the data and theory in the Egelstaff scattering law presentation $S(\alpha,\beta)$ per molecule vs α. For each initial energy, scattering law points for large scattering angles are to the right, and those for small scattering angles are to the left. In presentation of the data, the energy change is represented by $\beta = \hbar\omega/k_B T$, where $\hbar\omega$ is the neutron energy change, k_B is Boltzmann's constant, and T is the absolute temperature of the sample. Momentum change is represented by $\alpha = \hbar^2 Q^2/2Mk_B T$, where $\mathbf{Q} = \mathbf{k} - \mathbf{k}_0$, with $\mathbf{k}$ and $\mathbf{k}_0$ the final and initial wave vectors of the neutron, respectively. In this case the mass M was taken as 1 amu. The advantage of $S(\alpha,\beta)$ is that it is theoretically even in β.[6] Figure 6 shows the family of curves which are hand-drawn best fits to the data shown in Fig. 5.

As a check on the instrumental calibration, the partial differential cross sections for 0.0254-ev neutron energy were obtained from the $S(\alpha,\beta)$ curves in Fig. 6 and were integrated to obtain a total cross-section value. This value was 4.7% higher than that of Melkonian[2] and was considered to be in good agreement. The data are not normalized to Melkonian's value.

IV. THEORETICAL DISCUSSION

Krieger and Nelkin[5] have proposed that the inelastic scattering of neutrons with initial energies below the first vibrational state from simple molecules can be explained by assuming a continuum of rotational states, and by considering the zero-point vibrational states. In the present calculation the approach of Krieger-Nelkin was modified[7] to account for the effects of single vibrational state changes. To determine the vibrational states, a normal coordinate analysis of propane was made using a group vibrational method which is

[6] P. Schofield, Phys. Rev. Letters 4, 239 (1960).

[7] A. C. Zemach and R. J. Glauber, Phys. Rev. 101, 118 (1956), Eq. (3.22).
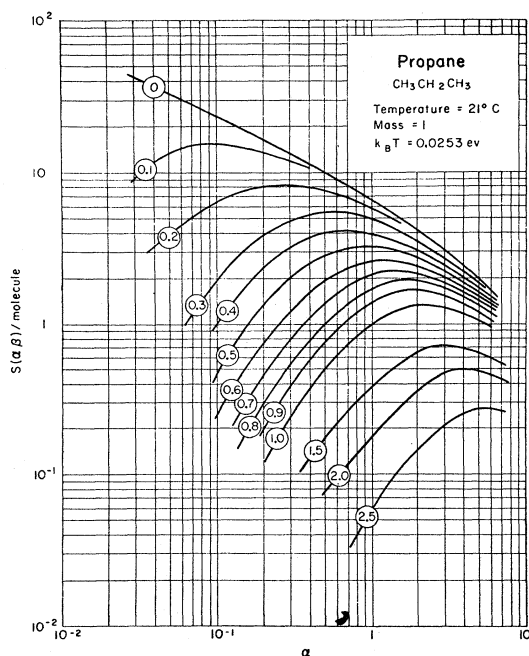
FIG. 6. Combined propane scattering law. These curves are hand-drawn best-fit curves to the data in Fig. 5. The circled numbers on each curve are the values of β.

weighted sums of the effects of the five types of atoms. The "ideal gas" curves in Fig. 5 are calculated using an average of the mass tensors for the three types of H atoms with neglect of direct carbon scattering and the vibrational factor. The Krieger-Nelkin curves of Fig. 5 used the average zero-point vibrational factor and the average H mass tensor.

The rotational energy constant for propane associated with the $J(J+1)$ energy term is 0.000033 ev. This is an order of magnitude lower than 0.00071 ev for methane. Thus, the classical rotational continuum approximation is adequate for propane. In methane the deviation from a continuum approximation has been calculated by Griffing[11] and shown to be most significant at low incident neutron energies and small scattering angles. In Fig. 1 at 16.3° the calculation underestimates the peak height and gives too large a width. This effect is seen in propane at $E_0=0.0101$ ev and $\theta=16.3°$ in Fig. 1 and at small α and β's of 0 and 0.1 in Fig. 5.

At small angles (except the $E_0=0.0101$ ev case), the calculation overestimates the cross section. This fact is

particularly suited to large molecules.[8] The symmetry group of propane has been shown experimentally by Lide[9] to be C_{2v}. The principal moments of inertia used were $I_x=66.44$, $I_y=58.24$, and $I_z=17.70$ amu A² which differ slightly from Lide's values. The force constants were taken from a variety of earlier work[10] on hydrocarbons except the torsional constant governing a methyl group's torsional oscillation about a C—C bond. Lide has given a lower limit of 2.7 kcal/mole for the threefold barrier and 3 kcal/mole or 0.13 ev is assumed for these calculations. Table I gives a comparison of all calculated and available experimental frequencies. The three lowest energy modes are of greatest importance in affecting scattering for the incident neutron energies considered in this experiment.

The assumption of direct scattering from the three types of H atoms and two types of C atoms was made together with single quantum changes of the three lowest vibrational modes and zero-point vibrational contributions from all modes. The spatial molecular orientation average was made analogous to Krieger-Nelkin. The mass tensors used for the translational and rotational parts of the scattering were 13.31, 12.66, and 12.73 amu for the three types of H atoms and 24.03 and 31.76 amu for the two types of C atoms. The modified Krieger-Nelkin curves of Figs. 1–5 are the

TABLE I. Propane vibrational levels.

Symmetry	Description[a]	Experimental[b] Freq. (cm⁻¹)	Experimental[b] Energy (ev)	Calculated Freq. (cm⁻¹)	Calculated Energy (ev)
A_1	CH₂ s. st.	2940	0.365	2930	0.364
	CH₃ u. st.	2940	0.365	3020	0.375
	CH₃ s. st.	2940	0.365	2880	0.358
	CH₂ s. def.	1460	0.181	1490	0.185
	CH₃ u. def.	1460	0.181	1470	0.183
	CH₃ s. def.	1380	0.171	1410	0.175
	CH₃ r., C₃ s. st.	1160	0.113	1180	0.146
	CH₃ r., C₃ s. st.	870	0.108	879	0.109
	C₃ s. def.	372[c]	0.046	367	0.046
A_2	CH₃ u. st.	· · ·		3020	0.375
	CH₃ u. def.	· · ·		1470	0.183
	CH₃ r.	· · ·		928	0.115
	CH₂ t.	· · ·		1200	0.148
	CH₃ t.	· · ·		312	0.036
B_1	CH₃ u. st.	2940	0.365	3020	0.375
	CH₃ s. st.	2940	0.365	2890	0.358
	CH₃ u. def.	1460	0.081	1480	0.184
	CH₃ s. def.	1380	0.171	1380	0.171
	CH₂ r., CH₃ u. st.	1340	0.166	1440	0.179
	CH₂ r., CH₃ u. st.	1050	0.131	973	0.121
	CH₃ r.	923	0.114	927	0.115
B_2	CH₂ u. st.	2940	0.365	3030	0.376
	CH₃ u. st.	2940	0.365	3020	0.374
	CH₃ u. def.	1480	0.153	1480	0.183
	CH₂ r.	748	0.093	795	0.099
	CH₃ r.	1190[d]	0.147	1200	0.149
	CH₃ t.	· · ·		277	0.034

[a] The abbreviations u. st., s. st., u. def., s. def., denote vibrations which are unsymmetrical stretching, symmetrical stretching, unsymmetrical deformation, and symmetrical deformation, respectively, within the groups indicated. The abbreviations r. and t. denote rocking and twisting motions of the indicated groups. The descriptions are only qualitative.
[b] H. L. McMurry and V. Thornton, J. Chem. Phys. 19, 1014 (1951).
[c] J. R. Nielsen, from unpublished Raman data on propane gas.
[d] M. Randic and D. M. Simpson, Trans. Faraday Soc. 56, 1 (1960).

[8] H. L. McMurry, J. Mol. Spectroscopy 3, 203 (1959); 6, 439 (1961).
[9] D. R. Lide, Jr., J. Chem. Phys. 33, 1514 (1960).
[10] T. Simanouti, J. Chem. Phys. 17, 848 (1949) and previous papers referenced here. F. Stitt, J. Chem. Phys. 7, 297 (1939).

[11] G. W. Griffing, Phys. Rev. 124, 1489 (1961).

attributed to the uncertainty in the experimental angular and energy resolution. If both of these effects were included in the theoretical treatment, they would reduce the peak height and broaden the theoretical cross-section curves, particularly at the small angles at large initial energies where the cross section peaks and varies most rapidly.

The fact that the vibrational "bumps" show up in the wings of the calculation and not in the experimental data of Figs. 2–4 (this effect is evident at $1 \leqslant \beta \leqslant 2$ in Fig. 5) can be due to the following reasons: (1) The torsional barrier height that we have assumed may be too high. (2) A more exact average over molecular orientations may be required. (3) A computation accounting for energy resolution will broaden the theoretical curves and may smear out the "bumps."

At the 0.0736- and 0.102-ev back angles, where the neutron interacts more violently with the molecule, the assumption of simple harmonic vibrational modes is not adequate for the modes with low frequencies (see Figs. 3–4). This may explain the experimental data points at large α's falling above the modified Krieger-Nelkin curve in Fig. 5.

V. SUMMARY AND CONCLUSIONS

In propane the effects of discrete rotational levels are evident at only the smallest initial neutron energy and scattering angle; the rotational continuum assumption is adequate over most of the experimental range. The considerations of single vibrational state changes in the theory give better agreement with the experimental results than either the "ideal gas" or Krieger-Nelkin formulas. Discrepancies still exist in $S(\alpha,\beta)$ at large α and large β due primarily to the uncertain methyl barrier height for the three lowest energy modes, to the harmonic oscillator approximation for these modes, and to the approximate molecular orientation averaging.

Straggling Effects on Resonant Yields

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(Received September 20, 1961)

The finite nature of energy loss processes for a charged particle in a material leads to a "ringing" effect in the thick-target yield curve. The effect is described and evaluated.

I. ORIENTATION

FOR orientation we consider a resonant nuclear reaction at an energy E_R, with width Γ, induced in a thick target by a beam of charged particles of energy E_0. We ask for the yield curve $Y(E_0)$. The cross section is supposed to be of the standard form

$$\sigma(E) = \frac{\sigma_0}{1+4(E-E_R)^2/\Gamma^2}. \quad (1)$$

The standard consideration goes as follows: for $E_0 \ll E_R$ the yield is zero, for E_0 well above E_R the particles are degraded through the resonance by their ordinary energy-loss processes, and the yield is

$$Y_0 = \frac{\pi \sigma_0 \Gamma N}{2(-dE/dx)_{E_R}}, \quad (2)$$

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per incident particle, where N is the density of reacting targets. The transition from zero to Y_0 occurs over a range of energy Γ around E_R, given exactly by the partial integrals of (1) from zero to E_0.

We propose to discuss here an extra effect arising from the fact that the energy losses are not continuous, but occur in discrete jumps. The maximum energy loss in a single event is approximately $4m/M$ of the energy of the incident (heavy) particle, and it is the existence of beams of particles with energy resolution better than this that makes these considerations interesting. The number $4m/M$ is approximately $1/500$ for incident protons, and resolutions five times better than this are achievable.

To show that there is an interesting effect, consider first the purely fictitious case in which *all* the energy losses are in jumps β, with a probability p per unit length, so that $-dE/dx = p\beta$. Then the average distance traveled per jump is p^{-1}, and the yield is given