

For a large, but finite, system one would expect good results from again using asymptotic distributions (in this instance the sum of growing and decaying asymptotic exponentials) as trial functions. However, in a small system the asymptotic distributions would not become established and their use as trial functions would probably lead to large errors. Since in this situation one would expect the directional and adjoint directional fluxes to be relatively flat (in space), a more appropriate trial function might be

$$\phi(z, \mu) = \sum_{n=0}^N \phi_n(\mu) z^n, \quad (31)$$

with an analogous adjoint trial function. In this case, one allows the variational method to determine the angular dependences of the trial functions. This general approach to transport problems has been discussed elsewhere⁵.

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⁵G. C. POMRANING and M. CLARK, Jr., *J. Nucl. Energy, A/B*, **18**, 191, (1964).

Neutron Scattering by Polyethylene*

FREQUENCY SPECTRUM

Crystalline polyethylene is known to be formed by very long, kinked and only weakly interacting chains of CH₂ radicals^{1,2}. Therefore its frequency spectrum is radically different from the one corresponding to a three-dimensional crystal. In fact, analyzing its normal modes of vibration in terms of plane waves $e^{i\vec{k}\cdot\vec{r}}$, the corresponding frequencies can only depend on the projection of the wave vector \vec{k} on a direction parallel to the chains (at least to the extent that the interaction between neighboring chains can be neglected). In other words, for any direction of \vec{k} , the frequency ω depends only on the phase difference θ between the

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¹TUNG PO LIN and J. L. KOENIG, *J. Molec. Spectr.*, **9**, 228 (1962).

²M. TASUMI and T. SHIMANOCHI, *J. Molec. Spectra.*, **9**, 261 (1962).

vibration of corresponding elements of two neighboring radicals of the same chain. This dependence, as calculated by Lin and Koenig¹, is shown in Fig. 1 where the nine different branches (corresponding to the three atoms in each radical) can be assigned to the following vibrational modes:

T	C-C-C-C torsion
B	C-C-C bending
S	C-C stretching
R	CH ₂ rocking
Tw	CH ₂ twisting
W	CH ₂ wagging
Ib	H-C-H bending
Ss	H-C-H symmetric stretching
As	H-C-H antisymmetric stretching.

Only two of these branches go to $\omega = 0$ for $\theta = 0$ and can thus be considered as acoustical modes. The remaining ones are optical branches. This again shows very clearly the difference with a three-dimensional crystal for which the number of acoustical branches always is three.

We shall consider only the optical part of Lin and Koenig's frequency distribution since the acoustical part is perturbed, at least near $\omega = 0$, by the weak coupling between neighboring chains. Because of this, at very low frequencies one must recover the typical ω^2 behavior of a Debye crystal. Since a frequency spectrum is defined as the fraction of modes per unit frequency interval, in our case we have

$$f(\omega) = c \sum_i \frac{d\theta_i(\omega)}{d\omega},$$

where c is a normalization constant and the sum is over all branches crossing the frequency ω . Hence $f(\omega)$ becomes singular at all those points where $\frac{d\omega}{d\theta} = 0$. A histogram of the spectrum obtained in this manner, but leaving out the C-C stretching branch as well as the acoustical branches, is shown in Fig. 2. The reason for leaving out the S

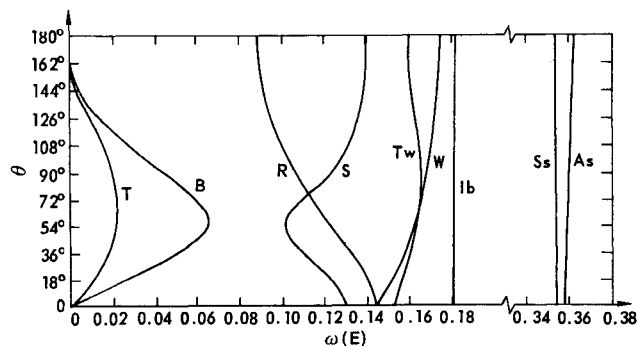


Fig. 1. Frequency dependence of the vibrational phase difference between neighboring CH₂ groups (Lin and Koenig).

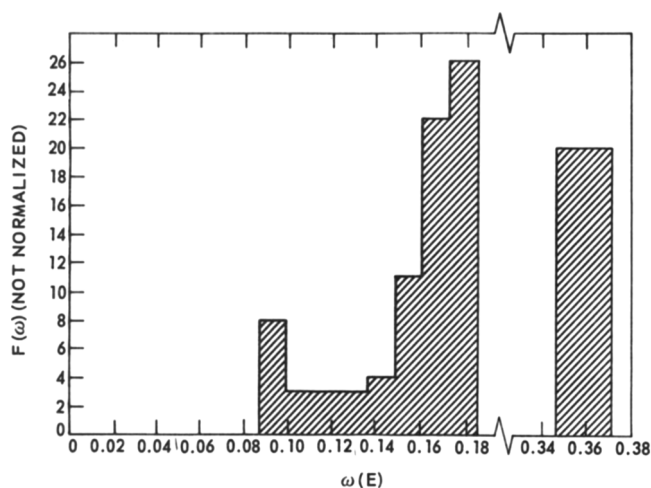


Fig. 2. Histogram of the frequency spectrum of polyethylene.

branch from the optical spectrum will become clear later.

NEUTRON SCATTERING KERNEL

From the frequency distribution described in the previous section it is possible, making reasonable assumptions, to derive a scattering kernel suitable to calculate thermal neutron spectra in polyethylene. Two different approaches to this problem have been attempted. First, the more accurate one, using a distributed frequency spectrum of the kind of Fig. 2, complemented by the adequately weighted C-C stretching contribution and a semiempirical acoustical spectrum given by Wunderlich³. There is need of a relative weighting of the different contributions to the effective frequency spectrum because, in the acoustical and S-modes, the CH₂ radicals move essentially as a whole unit of mass 14, whereas in the remaining optical modes the C-atoms are nearly at rest. The weighting factors, derived from the assumption that the CH₂ radicals remain rigid in the acoustical and S-modes, are $\frac{2}{15}$ for these and $\frac{13}{15}$ for the other modes. The scattering kernel obtained from this effective frequency spectrum gave neutron spectra in reasonable agreement with experiment^{4,5}, but perhaps because of numerical difficulties it was not possible to get a good fit to the experimental total scattering cross section.

While the calculation with the distributed frequency spectrum will still be pursued with improved methods, another approach was tried and found fairly successful. The idea is to lump the

frequency spectrum of Fig. 2 into a small number of discrete oscillators with weights proportional to the equivalent partial areas of the distributed spectrum. This situation can then be easily handled by the code⁶ GAKER, originally written for Nelkin's H₂O model, if one makes the additional assumption that the modes not included in the spectrum of Fig. 2 can all be treated in the short collision approximation, with T_{eff} equal to room temperature and a total weight of $\frac{1}{14}$ (corresponding to free translation of CH₂ radicals of mass 14).

This procedure is, in principle, the same as used by Goldman⁷ but the frequencies and weights obtained for the oscillators are quite different. In particular Goldman's highest frequency of 0.533 eV is in conflict with Lin and Koenig's theory since their hardest optical branch does not exceed 0.36 eV.

The number of discrete oscillators used in the present calculations was four and the weights and frequencies are given in Table I.

The numerical results obtained with these parameters seem to be in quite good agreement with experiment. In Fig. 3 we compare our calculated total scattering cross section with Bach's⁷ experimental data. Also shown in the figure is Goldman's calculation, and it is seen that the fit of the present model is quite satisfactory.

Furthermore the agreement obtained between calculated and measured neutron spectra is also quite good. This is seen in Figs. 4 and 5 which present two infinite medium spectra, one in 1% borated polyethylene (5.74 barns/H-atom) and the other one in paraffin containing enriched UF₄ with atom ratios $\frac{H}{U} = 189$ and $\frac{U^{235}}{U^{238}} = 0.02$.

Goldman's results for the borated polyethylene are also shown for comparison. The experimental points were taken from Refs. 5 and 8.

In summary, a neutron scattering kernel for

TABLE I

Frequencies and Weights Obtained for the Oscillators

Frequencies	Weights $\left(\frac{1}{M}\right)$	M
0.089	0.07746	12.91
0.14	0.2320	4.31
0.174	0.3096	3.23
0.360	0.3096	3.23

⁶H. C. HONECK, BNL-5826, (1961).

⁷D. T. GOLDMAN and F. D. FEDERIGHI, *Nucl. Sci. Eng.*, **16**, 165 (1963).

⁸J. C. YOUNG *et al.*, *Trans. Am. Nucl. Soc.*, **7**, 1 p. 71, (June, 1964).

³B. WUNDERLICH, *J. Chem. Phys.*, **37**, 1207, (1962).

⁴D. E. PARKS, unpublished.

⁵J. C. YOUNG *et al.*, *Nucl. Sci. Eng.*, **18**, 376 (1964).

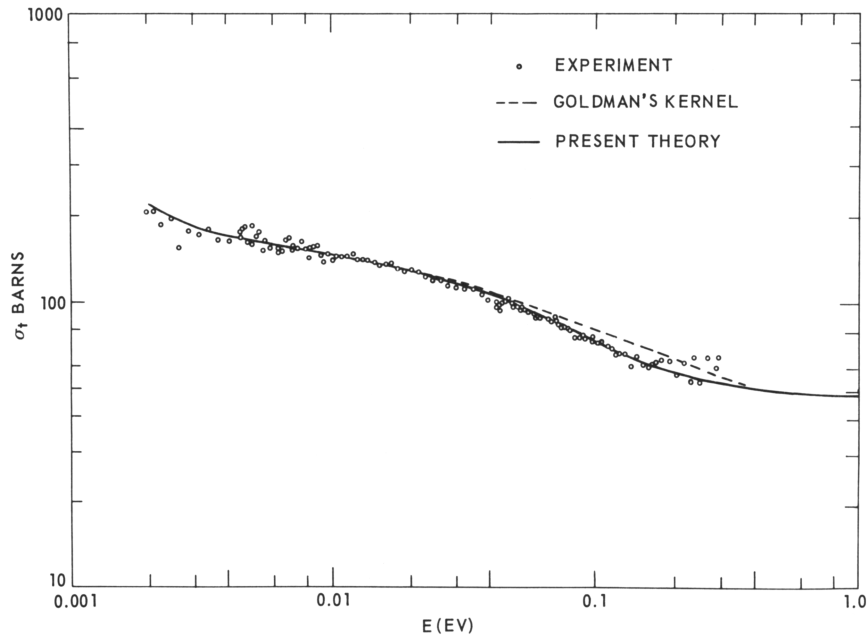


Fig. 3. Total cross section of polyethylene.

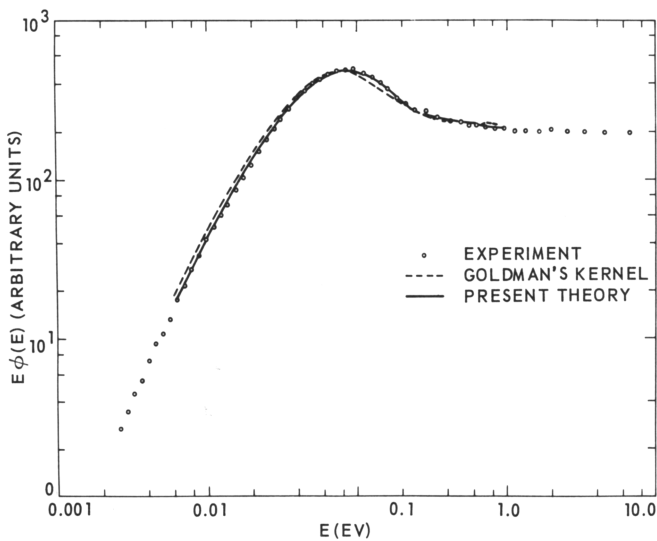


Fig. 4. Infinite medium neutron spectrum in 1% borated polyethylene.

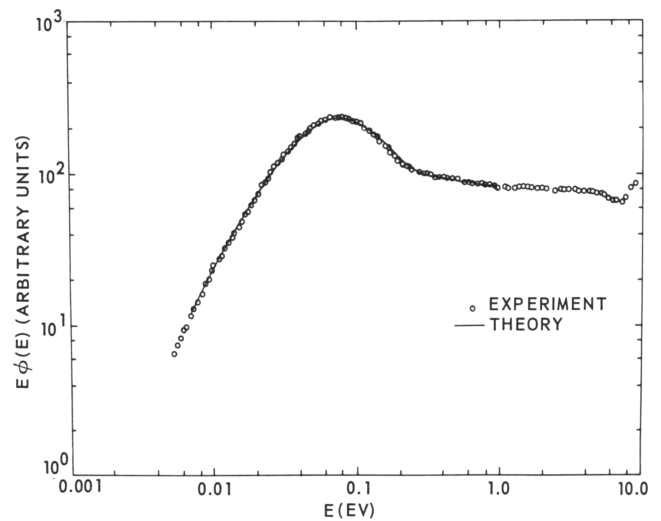


Fig. 5. Infinite medium neutron spectrum in paraffin containing enriched UF_4 .

polyethylene has been calculated lumping a theoretical frequency spectrum given by Lin and Koenig into four discrete Doppler-broadened oscillators. The total scattering cross section and neutron spectra in different media predicted by this model are in good agreement with experiment.

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NOTE ADDED IN PROOF:

Somewhat better agreement with recent double differential cross-section measurements in the energy range 0.15-0.50 eV (Kirouac *et al.*, Rensselaer Polyt. Inst. Progress Report Nov. 1964) was obtained by splitting the spectrum of Fig. 2 into only 3 lines at 0.09, 0.17 and 0.36 eV with weights of 0.10, 0.49 and 0.34 respectively.