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Liquid and Amorphous Total Scattering Instrument at KENS
- HIT -

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Abstract

Liquid and Amorphous Total Scattering Instrument (HIT) at KENS is in operation since 1980. It was originally designed to measure the S(Q) of liquids and amorphous solids composed of heavy nuclei with high count rate and modest resolution of the momentum transfer Q. Recently proposals which desire to measure the S(Q) of materials composed of light nuclei or with better resolution are increasing in number. The effects of the resolution and the inelastic scattering on observed S(Q) are, therefore, reexamined using machine parameters of the HIT. Current performance of the HIT is briefly reported as well.

1. Introduction

The Liquid and Amorphous Total Scattering Instrument (HIT) at KENS was constructed in 1980. Since then it has been operated for more than 5 years. During this period many users from outside universities performed measurements of S(Q) on various kinds of liquids and amorphous materials. Many scientific results have been obtained using this machine and significant contributions to the structure determination of liquids and amorphous solids have been made.

The HIT machine was originally designed to measure the S(Q) of liquids and amorphous solids with high count rates and modest resolution of momentum transfer Q.[1] The target materials we supposed were those composed of heavy nuclei or those of which inelastic scattering is not significant.

Recently proposals to measure the S(Q) for the materials containing light nuclei or with better resolution are increasing in number at KENS. Therefore we reexamined the effects of the resolution and the inelastic scattering on observed S(Q) using the machine parameters of the HIT. In this report we summarize the current performance of the HIT machine briefly and discuss how the resolution and the inelastic scattering affect the observed S(Q).

2. Outline of the machine

A schematic drawing of the current HIT machine is shown in Fig. 1. The machine views the ambient temperature solid polyethylene moderator. A sample is located 5,053 mm from the moderator. The evacuated sample chamber is 450 mm in diameter so that it has enough room to install a sample changer and sample enviromental equipments such as a high temperature furnace, low temperature refrigerator, magnet and so on. The neutron flux at the sample position is about 5×10^5 n/cm^2/sec/2μA in the range of 0.3 to 0.7 Å which we use mainly for S(Q) measurement.

The neutron detectors used are He-3 gas counters (1/2" in diameter, 12" in active length and 20 atm in filled pressure). In the high angle region (150° and 90°) the detectors are located at geometrical time focusing positions, while in the lower angle region they are located at electrical hardware time focusing positions described later in detail. The scattered flight path length has been determined to give both high

count rates and modest geometrical Q resolution. The scattering angle and the flight path length for each detector now used are listed in Table 1.

The electrical hardware time focusing system which consists of a CAMAC module was newly developed.[2] The block diagram is shown in Fig. 2. The new system is the same in principle as the time focusing one using a computer. The main difference between the two systems is that the former system does not need any computing time so that it can fit measurements with the higher count rate. In this system each detector has own time analyser, of which the time width $\Delta\tau$ is chosen easily by a software to satisfy the following time focusing condition.

$$\Delta\tau = \frac{L \sin\Theta}{L_r \sin\Theta_r} \Delta\tau_r , \qquad (1)$$

where L and 2Θ are the total flight path length and the scattering angle, respectively. Subscript r denotes the reference detector. The all time analysers (sub modules in Fig. 2) involved in a certain detector bank are controlled by one main module. The counts accumurated in a sub module during time interval $\Delta\tau$ are added to the stored counts in the corresponding address in Q-space of the main memory giving a time-focused result. The system is controlled by a personal computer PC 9800 (NEC). Acceptable maximum count rate is expected to be about 6 counts/μs per each time analyser.

3. Geometrical resolution

The resolution in Q space was estimated by a Monte Carlo technique. The geometrical sizes of moderator (100 x 100 mm^2), sample (8 mm in diameter and 40 mm in height as a typical case) and detector (1/2" in diameter and 4 ∿ 12" in length depending on scattering angles) were taken into account. The time structure of the incident neutron pulse was included in an approximate manner as well. The resolution ΔQ was defined as a root mean square deviation of the Q value from the mean value $\bar{Q}$, i.e. $\Delta Q = \langle (Q - \bar{Q})^2 \rangle^{1/2}$. The calculated results are shown in Fig. 3. The broken lines show the case of the poor resolution, while the solid lines do the case of the good resolution. In the latter a limitted detector height was assumed except for 90° detectors, while in the former a slightly off time focusing position was chosen for 90° detector bank.

Usually we utilize the neutrons only in the wavelength range of 0.3 - 0.7 A for the S(Q) measurement, except for the low Q measurement with low angle counter, in order to avoid the serious distortion of S(Q) arising from non-constant Q scan of the instrument. The range between the two dots on each curve in the figure corresponds to this wavelength range. Figure 4 demonstrates the effect of the geometrical resolution on the simulated S(Q) for a model system. The result suggests that the resolution ΔQ must be at least 0.2 A^{-1} or less if one requires the precise measurement of S(Q).

4. Effect of the inelastic scattering

The inelastic scattering is a serious problem in the S(Q) measurement when a sample contains light atoms. In order to know how the inelastic scattering affects observed S(Q) at high Q, we simulated experiment on the HIT using the following simple models of $S(Q,\omega)$'s.

$$S_{self}(Q,\omega) = \left[\frac{M}{2\Pi\hbar^2Q^2kT} \right]^{1/2} \exp\left[- \frac{M}{2\hbar^2Q^2kT} \left(\hbar\omega - \frac{\hbar^2Q^2}{2M}\right)^2 \right],$$

$$S_{int}(Q,\omega) = \left[\frac{M^*}{2\Pi\hbar^2Q^2kT} \right]^{1/2} \exp\left[- \frac{M^*}{2\hbar^2Q^2kT} \left(\hbar\omega - \frac{\hbar^2Q^2}{2M^*}\right)^2 \right] \cdot (S(Q)-1) \quad , \tag{2}$$

where M is the atomic mass and M^* is the effective mass for the coherent scattering just for convenience. This model is enough to examine the effect of the inelastic scattering on S(Q) in high Q region although it is not in low Q. In the simulation an incident neutron spectrum $\Phi(E)$ was approximated by the following analytical form,

$$\Phi(E) = \Phi_o \left[\Phi_{th} \left(\frac{E}{E_T^2} \right) \exp\left(- \frac{E}{E_T}\right) + \Phi_{epi} \frac{\Delta(E)}{E^{0.878}} \right], \tag{3}$$

where E_T = 35.2 meV, Φ_{th}/Φ_{epi} = 6.89 and $\Delta(E) = 1/[1 + (5E_T/E)^7]$.
The detector efficiency $\eta(\lambda)$ was approximated by the following equation,

$$\eta(\lambda) = 1 - \exp(a + b\lambda + c\lambda^2) \qquad (0.05 < \lambda < 9.6\ \text{A}) , \qquad (4)$$

where a = 0.01839, b = -1.44457 and c = 0.007479. Figure 5 shows the instrumental integration paths calculated for the HIT machine. The integration of $S(Q,\omega)$ with respect to ω is carried out by the HIT along these integration paths with weighted factors of Φ (E) and $\eta(\lambda)$. S(Q)'s simulated for M/m = 100 and M^*/m = 200, where m is a neutron mass, are shown in Fig. 6. The effect of the inelastic scattering on the simulated S(Q) is not serious as expected for samples with heavy mass. On the other hand for samples with light mass such as M/m = 11 and M^*/m = 27 the effect is serious as shown in Fig. 7. The following three effects on the S(Q) due to inelastic scattering may be confirmed :

i) deviation of $S_{self}(Q)$ from the unity,

ii) damping of the oscillation of S(Q), and

iii) phase shift of the oscillation of S(Q).

All of three effects are significant at higher scattering angle. We have often encountered these three effects in actual experiments. Among these three effects, the second and the third are very important for structural investigations.

Powles[3] proposed a correction method for the inelastic scattering for various molecules, but it seems hard to make precise corrections when a sample contains three or more chemical species. The materials we are interested in are often multi-component systems, such as aqueous solutions. Therefore, we are planning to install a new detector systems at the lower scattering angles (less than 30°) using ^{6}Li glass scintillators. In order to measure the S(Q) in the range of Q over 30 A^{-1} or more with a reasonable resolution, the momentum resolution must be less than 0.2 A^{-1} in the whole Q range. To achieve this required resolution and to still keep good count rate, a coarse converging collimator may be useful, and the glass scintillators will be arranged as many as possible along the Debye-Scherrer cone on a focusing sphere of the converging collimator. Design work to improve the HIT is in progress.

Acknowledgement

The authors would thank Mr. S. Satoh for providing the new time analyser system including computer software. Their thanks are also to all members of the HIT group.

References

1) N. Watanabe, T. Fukunaga, T. Shinohe, K. Yamada and T. Mizoguchi, Proc. of ICANS-IV, KENS Report II (1981), 539.

2) S. Satoh, KEK Internal 85-8 (1985) (in Japanese).

3) J. G. Powles, Molec. Phys. 36 (1978), ibid 37 (1979) 623.

Table 1. Parameters of the HIT machine

(Scattered flight path length at detector center L_2, scattering angle at detector center 2θ and time width of time analyser ΔT)

Detectors	L_2 (mm)	2θ (degree)	ΔT (μsec)	Note
150° bank				
150 a2	301.6	155.3		
150 a3	327.0	152.5	2.0	geometrical
150 a4	349.9	150.0		time focusing
90° bank				
90 a1	255.5	93.6		
90 a2	330.0	90.2	2.0	geometrical
90 a3	418.3	87.6		time focusing
50° bank				
50 b1	514.1	42.0	4.0	electrical hardware
50 b4	500.5	47.8	4.5	time focusing
30° bank				
Fa 15	586.3	26.2	4.0	electrical hardware
Fa 18	557.0	31.5	4.75	time focusing
20° bank				
Fa 11	631.1	20.0	8.0	electrical hardware
Fa 13	607.17	23.0	9.125	time focusing
13°				
Fa 6	697.6	13.3	8.0	
7°				
Fa 6	779.4	7.4	8.0	

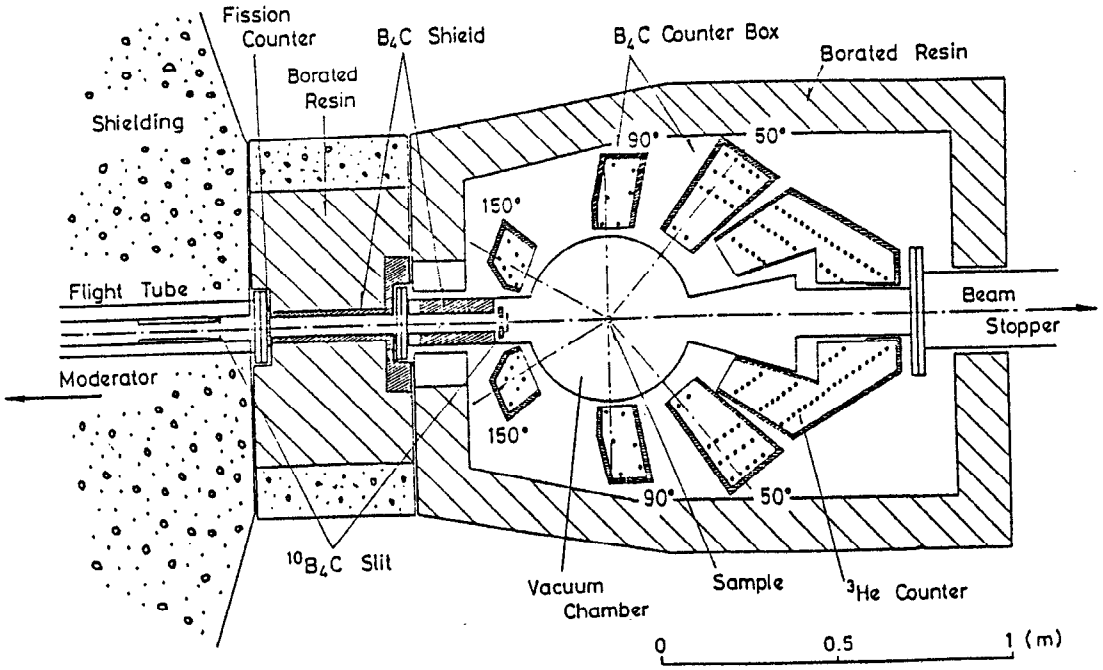

Fig. 1. A schematic drawing of the current HIT machine.

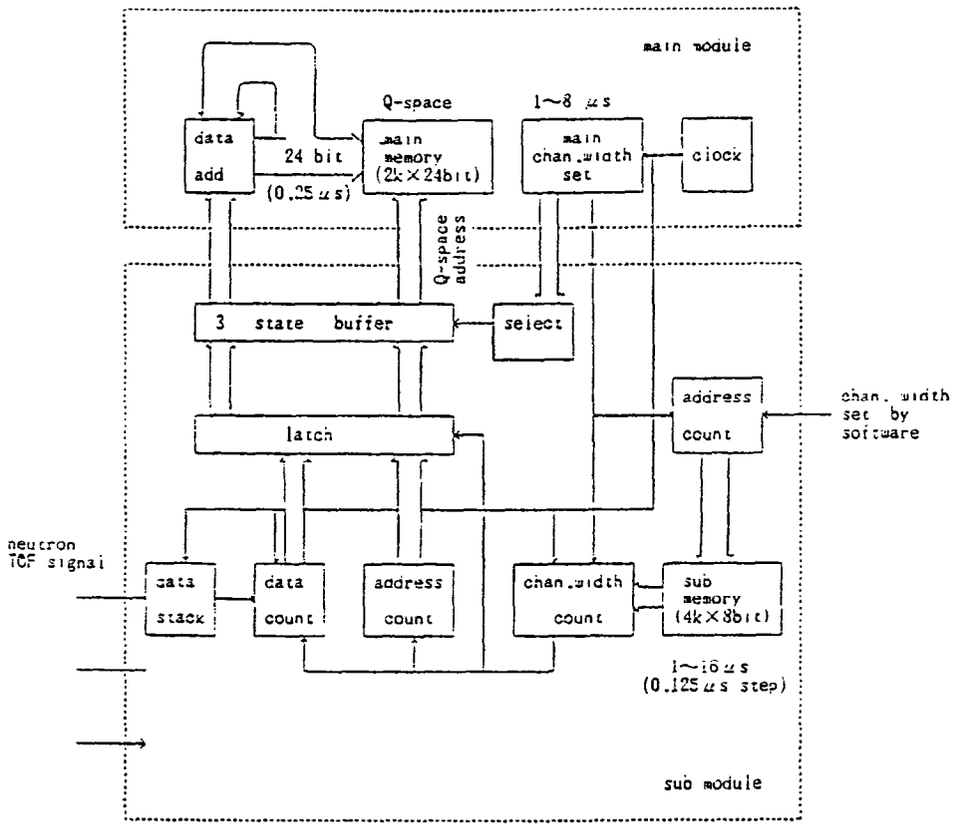

Fig. 2. Block diagram of CAMAC module for hardware time focusing.

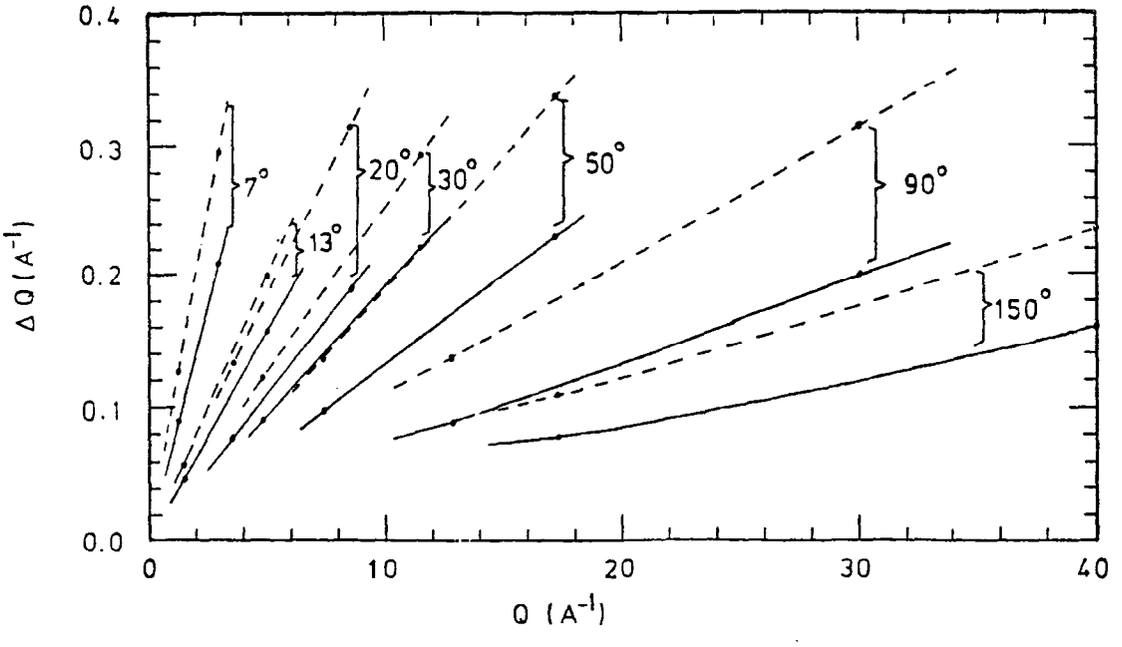

Fig. 3. Geometrical resolution of momentum transfer Q calculated for various detector banks now used in the HIT. Broken lines show poor resolution, while solid lines good resolution. The region between two dots on each line corresponds to the wavelength between 0.3 and 0.7 Å.

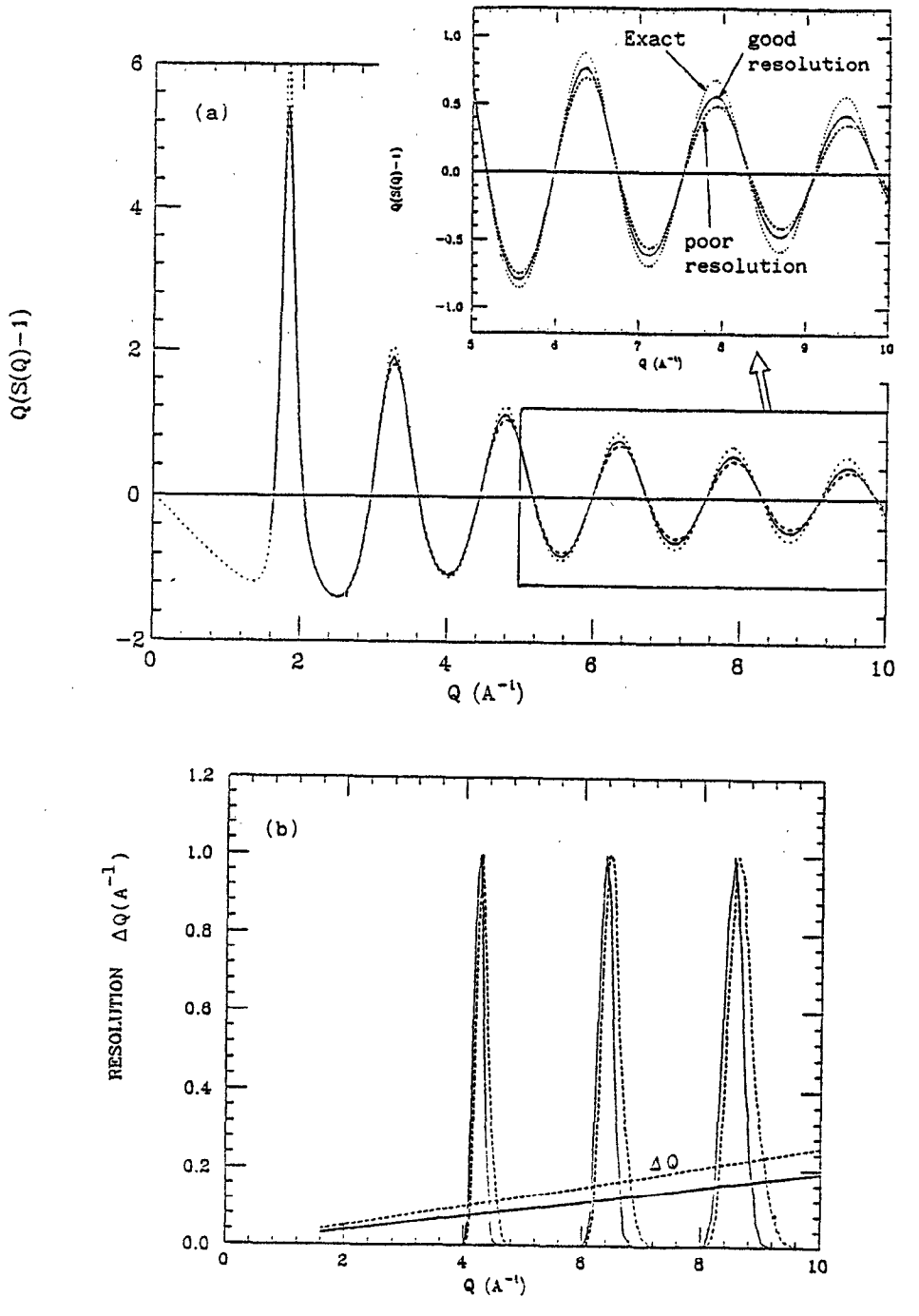

Fig. 4. Effect of the Q resolution on S(Q).
(a) The dotted curve is an exact S(Q) calculated for hard sphere system. Broken and solid curves are the S(Q)'s calculated with the poor and good resolutions shown in the lower figure.
(b) A resolution ΔQ for $2\Theta = 30°$ which is the same as already shown in Fig. 3. Solid line shows good resolution, while broken line poor resolution. Resolution functions at ca. 4.2, 6.4 and 8.5 A^{-1} are shown as well.

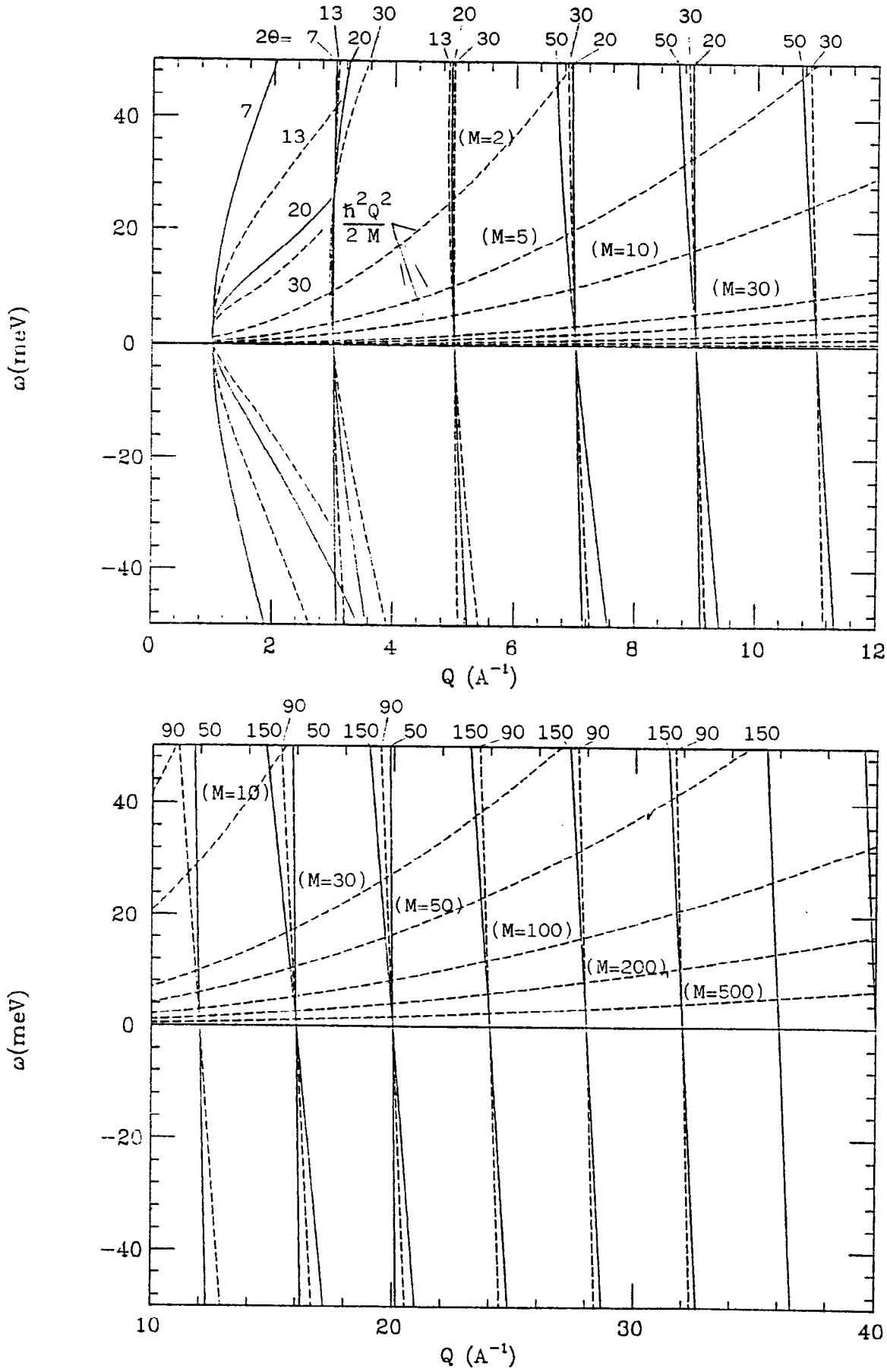

Fig. 5. Instrumental integration paths of the HIT. Curves of recoil energy $\hbar^2Q^2/2M$ for various M's are also shown.

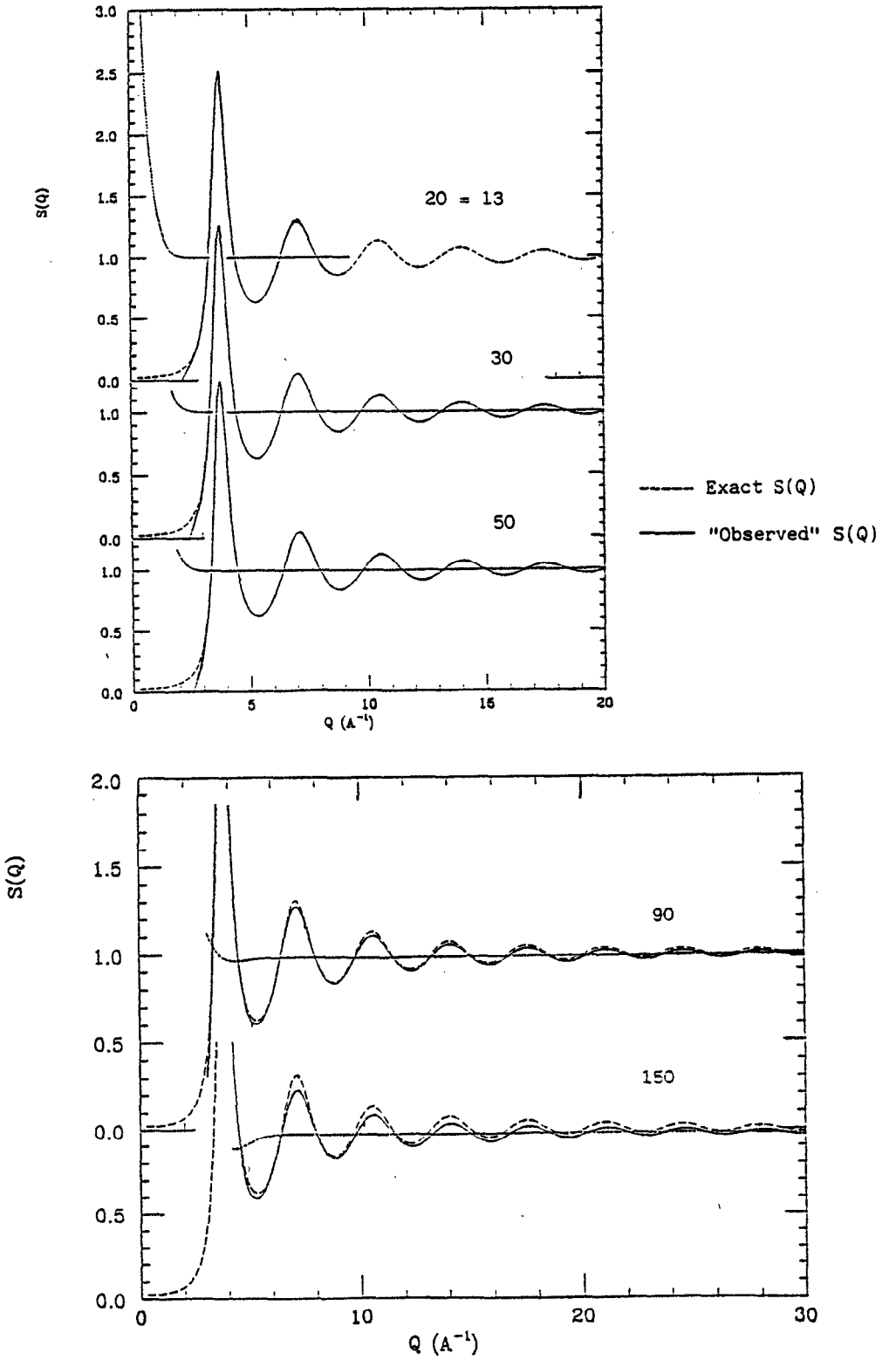

Fig. 6. The comparison of S(Q)'s simulated for heavy nuclei (M/m = 100, M^*/m = 200, solid curves) and exact ones (broken curves).

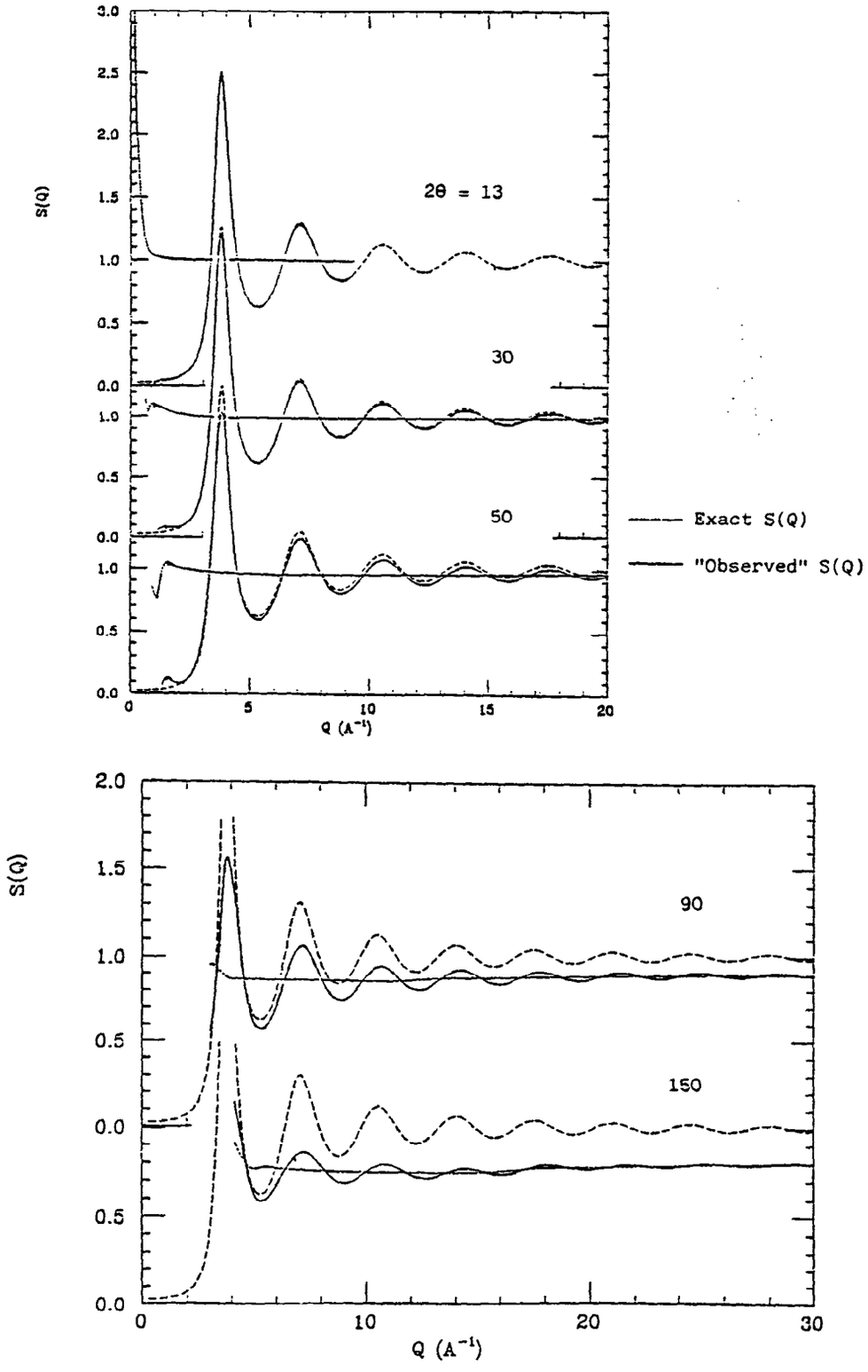

Fig. 7. The comparison of S(Q)'s simulated for light nuclei (M/m = 11, M^*/m = 27, solid curves) and exact ones (broken curves).