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Crystal Analyzer Type Spectrometer LAM-D at KENS Spallation Thermal Neutron Source

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We constructed the inelastic scattering neutron spectrometer LAM-D covering the energy transfer range up to 300 meV with the resolution $\Delta\varepsilon/E_1$ less than 6%. The basic princile of LAM-D is the same as the quasielastic scattering spectrometers LAM-40 and LAM-80 at KENS.

Introduction

In the previous paper [1], we have reported two crystal analyzer type quasielastic neutron scattering spectrometers LAM-40 and LAM-80 which have been installed at the KENS spallation cold neutron source at the National Laboratory for High Energy Physics (KEK), Tsukuba.[2] LAM-40 with PG analyzer mirrors can cover the energy transfer range up to 10 meV with a conventional energy resolution $\Delta\varepsilon$=0.2 meV at the elastic position. LAM-80 with PG mirrors has a rather high energy resolution of $\Delta\varepsilon$=0.02 meV while the energy transfer range is limited below 0.5 meV. Applying mica crystals, we have recently made a great progress on LAM-80 in the energy resolution. This new machine LAM-80ET has been reported elsewhere.[3]. The energy transfer ranges covered by the two spectrometers are not so narrow compared with other machines which have the same order of the energy resolution.[4] However, it is often essential to measure in a wide energy range up to several hundred meV for some physical phenomena such as molecular vibrations. At the present stage, it is probably impossible to design a single quasi- and inelastic spectrometer covering such a wide energy range with high energy resolution. Therefore, in addition to LAM-40 and LAM-80, we have designed and constructed an inelastic scattering spectrometer (LAM-D) to cover the energy range up to 300 meV with the energy resolution of $\Delta\varepsilon/E_1$

less than 6% through the whole energy range. The basic principle of LAM-D is the same as LAM-40 and LAM-80 while some special considerations have been made on the design of LAM-D for the high energy transfer measurements. Prototype of LAM-D was installed at the KENS thermal neutron beam hole (H-6) in 1982 and the final version is now in practical use on the thermal beam hole (H-9). Combination of the three spectrometers LAM-D, LAM-40 and LAM-80 is very powerful for investigation of dynamics of various molecular systems.

In this paper, we will first recall the basic principle of the LAM-type spectrometer and clarify the characteristics of LAM-D comparing with LAM-40 and LAM-80 in sec.2 and the description and the performance of LAM-D will be given in secs. 3 and 4 including some experimental results.

2. Basic Principle of LAM-type Spectrometer

General layout of the LAM-type spectrometer using a pulsed neutron source is illustrated in Fig.1, which is an inverted geometry quasi- and inelastic spectrometer. Pulsed neutrons with a wide energy distribution emitted from the thermal or cold moderator (H_2O at room temperature and CH_4 at 20K for thermal and cold moderators, respectively, at KENS) are incident on a sample after flying through the first flight path (l_1) and the scattered neutrons with a fixed energy are selected by analyzer mirrors and detected by neutron counters. The distance from the sample to the analyzer mirror is the same as that from the mirror to the detector, which is a half of the second flight path length (l_2). Measurement of energy transfer of neutrons is performed by a time-of-flight (TOF) technique. In the case of TOF measurements, the energy resolution required is attained by selecting an appropriate length of neutron flight path. The energy resolution of the crystal analyzer mirror can be selected by adjusting the Bragg angle. The combination of devices enabled us to design a set of quasi- and inelastic spectrometer with large flexibility of performance.

For the LAM-type spectrometer, the time spectrum of the scattered neutrons from the sample at a scattering angle θ is given by [1,5]

$$\eta(t,\theta) = \mathrm{const}\iint\phi[E_1, t - l_2/(2E_2/m)^{1/2}]\,\sigma(E_1 \rightarrow E_2, q)R(E_2)dE_1dE_2 \quad (1)$$

where $\phi(E_1,t_1)$ is the incident neutron flux at time t_1 into the sample, $\sigma(E1 \rightarrow E2,\theta)$ is the incoherent differential scattering cross-section, $R(E_2)$ is the energy resolution function of the crystal analyzer mirror, m is the neutron mass, and l_2 is the average flight path length of the scattered neutrons.

The quantities measured are the energy transfer ε and the scattering vector **Q** defined by

$$\varepsilon = E_1 - E_2 \tag{2}$$

and

$$\mathbf{Q} = \mathbf{k}_1 - \mathbf{k}_2 \tag{3}$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave number vectors of the incident and the scattered neutrons, respectively and Q^2 is given by

$$Q^2 = (2m/h^2)[E_1 + E_2 - 2(E_1E_2)^{1/2}\cos\theta\,] \tag{4}$$

The energy resolution of the LAM-type spectrometer is given by [1,5]

$$\Delta\varepsilon = 2E_1[(\Delta t_z^2 + \Delta t_1^2 + \Delta t_2^2)/t_1^2 + (E_2/E_1)^2\{(\cot\theta_B\Delta\theta_B)^2 + (\Delta\tau/\tau)^2\}]^{1/2} \tag{5}$$

where Δt_z is the pulse width at half-maximum dependening on the neutron energy and Δt_1 and Δt_2 are the variances of the flight time of neutrons due to the fluctuations of the flight path length l_1 and of the analyzed energy E_2, respectively. θ_B is the Bragg angle of the analyzer mirror and $\Delta\theta_B$ is the angular spread of the Bragg angle. $\Delta\tau$ is the variance of the reciprocal lattice vector τ of the analyzer crystal due to the limitation of the number of lattice planes contributing to the reflection. [5] The first, second and third terms in the right hand side of eq.(5) are the variances in the time-fo-flight measurement and the forth and fifth terms are due to the analyzer mirror resolution. The first term $\Delta t_z/t_1$ was calculated as a function of the incident neutron energy E_1 and the result is shown in Fig.2. In this calculation the first flight path length was taken to be 4.3 m. This term decreases with increasing the incident energy E_1 because the pulse width of neutrons Δt_z decreases with increasing E_1. In the case of high resolution quasielastic machines such as LAM-80, this contribution to the energy resolution is serious so that long flight path is essential to diminish it. The second term which is determined by the sizes of the neutron beam and the sample is small enough compared with other terms in our case as shown in Fig.2. The third term $\Delta t_2/t_1$ was also calculated as a function of E_1, taking l_1 and l_2 as 4.3 m

and 0.6 m, respectively, and the average analyzing energy as 4.4 meV. In contrast to the first term, it increases with E_1. In the case of inelastic spectrometers covering high energy region like LAM-D, this contribution should be diminished without sacrificing the other contributions to the resolution. This term can be rewritten as

$$\Delta t_2/t_1 = (1/2)(l_2/l_1)(E_1/E_2)^{1/2}(\Delta E_2/E_2) \qquad (6)$$

According to eq.(6), in order to achieve higher energy resolution in the high energy region, it is necessary to decrease the second flight path length l_2. Therefor, l_2 of LAM-D was designed to be 0.54 m, which is much shorter than that of LAM-40 and LAM-80 (1.2m). It is necessary to evaluate the spread of the Bragg angle $\Delta\theta_B$ for calculation of the forth term. The resolution function of the analyzer mirror $R(\theta_B)$ can be calculated by the following equation [5],

$$R(\theta_B) = \iiint p(r,\Sigma,S)\ |r-\Sigma|^{-2}\ |\Sigma-S|^{-2} \times \delta[\ \theta_B - \hat{\theta}_B(r,\Sigma,S)\ ]\ dr d\Sigma dS \qquad (7)$$

where $\hat{\theta}_B(r,\Sigma,S)$ is the Bragg angle determined by the positions on the sample r, the mirror Σ and the counter S, and $p(r,\Sigma,S)$ is a factor representing efficiency due to the projection of each plane and the mosaicness of the crystals. The integrations are performed with respect to the volume of the sample, the surface of the mirror and the surface of the counter window. $R(\theta_B)$ calculated for the Bragg angle 39 deg is shown in Fig.3. In the calculation, the size of the crystal pieces is 12 mm x 12 mm and 2 mm in thickness with 1.2 deg mosaicness and the total number of the pieces is 56; they are arranged in seven columns. The average second flight path length was taken to be 0.6 m and the cylindrical sample is 14 mm in diameter, 0.1 mm in thickness and 60 mm in height. The window size of the counter is 14 mm in width and 60 mm in height. The spread of the Bragg angle was determined to be 1.1 deg from $R(\theta_B)$ in Fig.3. Using this value, the E_1 dependence of the forth term $(E_2/E_1)\cot\theta_B\Delta\theta_B$ was calculated in Fig.2. The contribution of this term is large in the low energy region while it is not so serious in the high energy region. In the case of LAM-D, the fifth term $(\Delta\tau/\tau)$ is small enough to neglect as compared with the other terms as shown is Fig.2.

Based on the results in Fig.2, we calculated the overall resolution of the spectrometer $\Delta\varepsilon/E_1$ through eq.(5). The results are shown as a function of the

incident energy E_1 in Fig.4 where $\Delta\varepsilon/E_1$ for l1=8.6 and 12.9 m are also indicated.

3. Description of the LAM-D Spectrometer

Fig.5 illustrates the configuration of the LAM-D spectrometer. The spectrometer is installed at the H-9 thermal neutron beam hole at KENS, which sees the thermal neutron moderator (H_2O at room temperature). Energy distribution of neutrons from the thermal beam hole is shown in Fig.6. The flight path from the moderator to the sample is evacuated and the length (l_1) is 7.25 m.

Each analyzer mirror is made from 56 pyrolitic graphite (PG) crystal pieces of 12 mm x 12 mm x 2 mm in size with a mosaic spread of 1.2 deg. As seen in Fig.5, two analyzer mirrors are symmetrically mounted at the scattering angle 35 deg and the other two at 85 deg. The 15 cm beryllium (Be) filters, which are cooled by liquid nitrogen, diminish the higher order reflections from the analyzer crystals. The length of the Be filter was determined by Monte Carlo simulation for transmission of neutrons with total atomic-cross section of both σ_t=0.5 and 7.5 barns, which correspond to those below and above the Be cutoff wavelength [6]. The results of the simulation is given in Fig.7.

The set of the mirrors and Be-filters is housed in an evacuated container surrounded by neutron shield made of borated resin and cadmium sheets. Moreover, each analyzer mirror has its own inner shield.

A hollow cylindrical shape was adopted for the sample container to assure identical geometrical conditions for every analyzer mirrors. The typical dimensions of the sample are 14 mm in diameter and 60 mm in height. These dimensions were determined by the optimization condition calculated between the energy resolution and the counting efficiency. The thickness of the sample depends on the scattering and absorbing cross section. It is essential to use a thin sample in order to avoid self-shielding and multiple scattering in the sample . For example, in the case of water, a sample of 0.1 mm thick is desirable. The outer and inner walls are made of aluminum of 0.25 mm in thickness. This container can be also used for measurements on the LAM-40 and LAM-80 spectrometers.

4. Performance

Raw spectra of vanadium measured with LAM-D are shown in Fig.8(a) and (b) for scattering angles of 35 deg and 85 deg, respectively. Each spectrum was obtained by summing up two spectra measured at the symmetrical position with the same scattering angle. The thickness of the vanadium sample is 2 mm and the measureing time is 8 hours. The peak counts are given in the figures which enable us to estimate the intensity performance of the LAM-D spectrometer.

Expanding the spectra in the inelastic region in Fig.8, inelastic scattering from phonons in vanadium is clearly observed. Fig.9 shows the spectrum of vanadium measured with LAM-D without the 15 cm Be-filter. The higher order reflections from the analyzer mirror are very strong in the inelastic region. For example, the second peak is about 5 times larger than the first reflection peak. It is because the intensity of incident neutron at 4.4 meV (elastic energy) is much weaker than that for the second and third reflection as shown in Fig.6. The results indicate that the 15 cm Be-filter works efficiently.

Full-width at hlaf-maximum (FWHM) of the elastic peak evaluated from vanadium spectrum is 0.40 meV, which agrees with the theoretical value in Fig.4. It means that the evaluation of the energy resolution through eq.(5) works very well in the low energy region. However, it is difficult to confirm whether the estimation of resolution in the high energy region is correct or not, since the energy resolution cannot be measured directly in the high energy region. As shown in Fig.2, the resolution is mainly governed by the term $\Delta t_2/t_1$ which is determined by ΔE_2 and by convolution of ΔE_2 and $\Delta t_2/t_1$ in high and low energy regions, respectively. In the low energy region, as shown above, the theoretical evaluation works well. This probably means that we can precisely calculate ΔE_2 and therefor it is expected that the evaluation of the resolution in the high energy region also works well.

Inelastic scattering spectra from trans-1,4-polychloroprene (PCP) at 10 K are shown in Fig.10 where the spectra from the container are also indicated after normalization by the monitor counts. The thickness of the sample is 0.1 mm and the measuring time is 8 hours. These raw spectra make us to conclude that the performance of LAM-D for the S/N ratio is extremely good up to 300 meV. The intense signal below 30 TOF channel is due to fast neutrons with energy higher than Cd cut-off energy. Fortunately, this fast neutron background does not contaminate the sample data below 300 meV. In the PCP spectra we can clearly recognized C-H stretching at 2950 cm^{-1} and some

sharp peaks which agree with the results of normal coordinate calculation for a single chain of PCP.[7]

5. Conclusion

The spectrometer LAM-D with combination of the pulsed thermal neutron source have demonstrated satisfactory performance for the measurements of inelastic scattering in the energy range up to 300 meV. The basic principle of LAM-D is the same as that of the quasielastic spectrometers LAM-40 and LAM-80, so that the data reduction methods developed for LAM-40 and LAM-80 can be applied to data of LAM-D with small modifications. With combination of the three spectrometers, we can cover the energy range form 1μeV to 300 meV. It is very powerful to investigate condensed matter physics.

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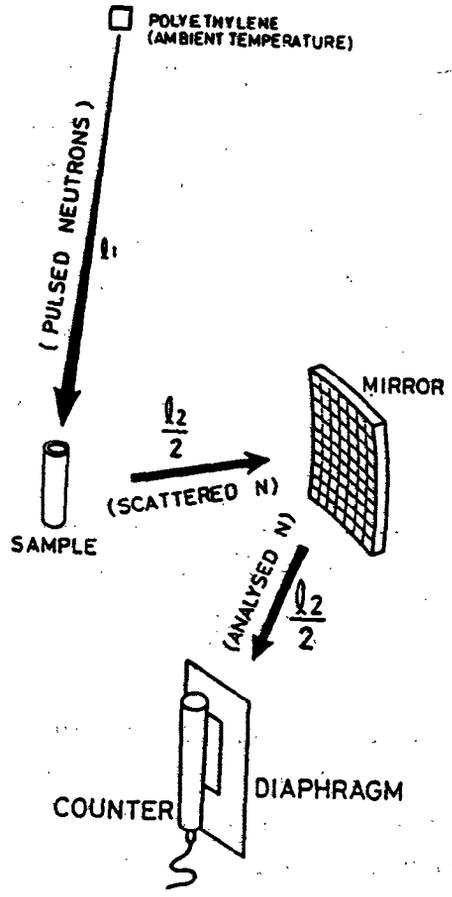

Fig.1. General layout of the LAM-type spectrometer using a pulsed neutron source. It is an inverted time-of-flight quasi- and inelastic scattering spectrometer.

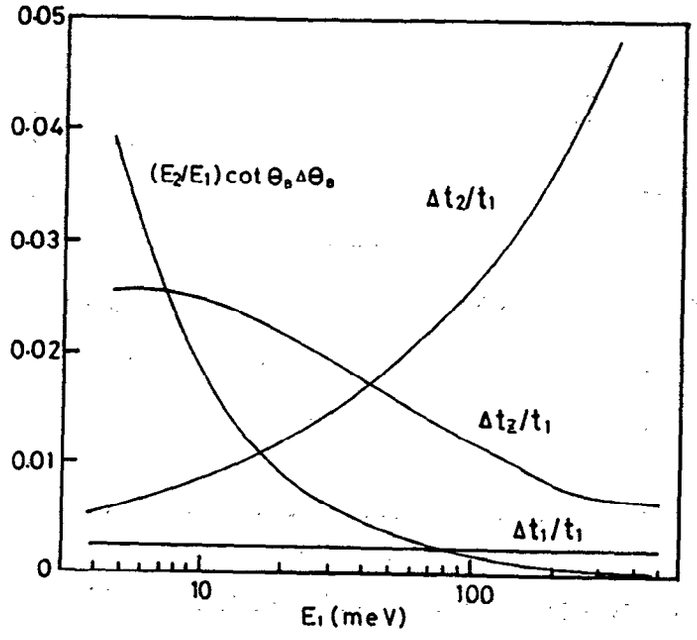

Fig.2. Incident energy dependence of each variance contributing to the enenrgy resolution [see eq.(5)].

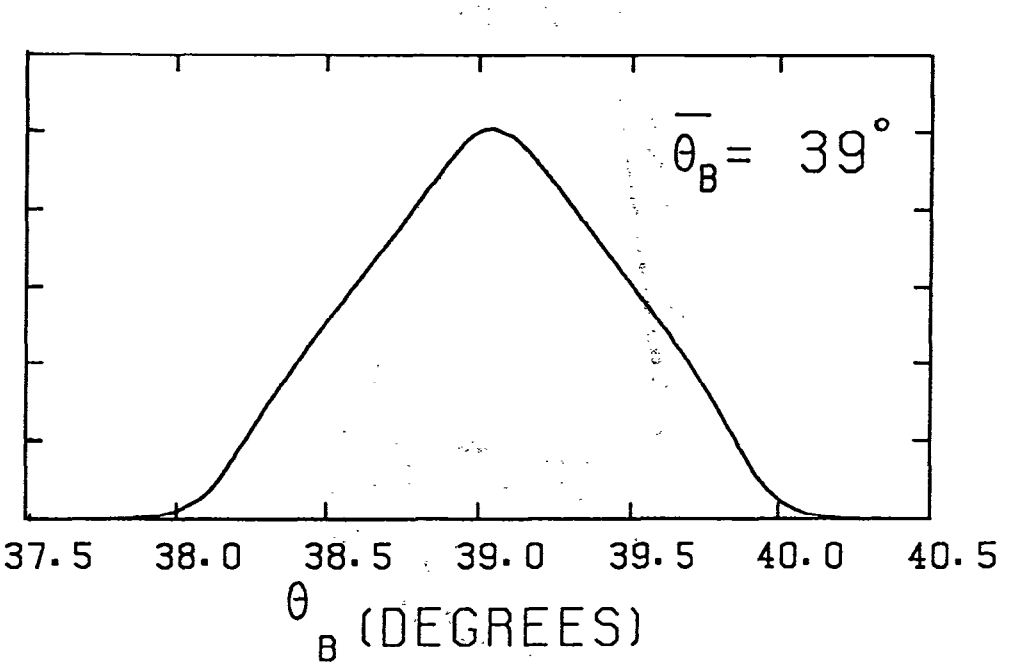

Fig.3. Calculated resolution function of the analyzer mirror for the Bragg angle 39 deg. The size of the sample is 14 mm in diameter, 0.1 mm in thickness and 60 mm in height. The number of the pieces of PG crystals of 12 mm x 12 mm x 2 mm in size is 56. The size of the counter window is 14 mm in width and 60 mm in height.

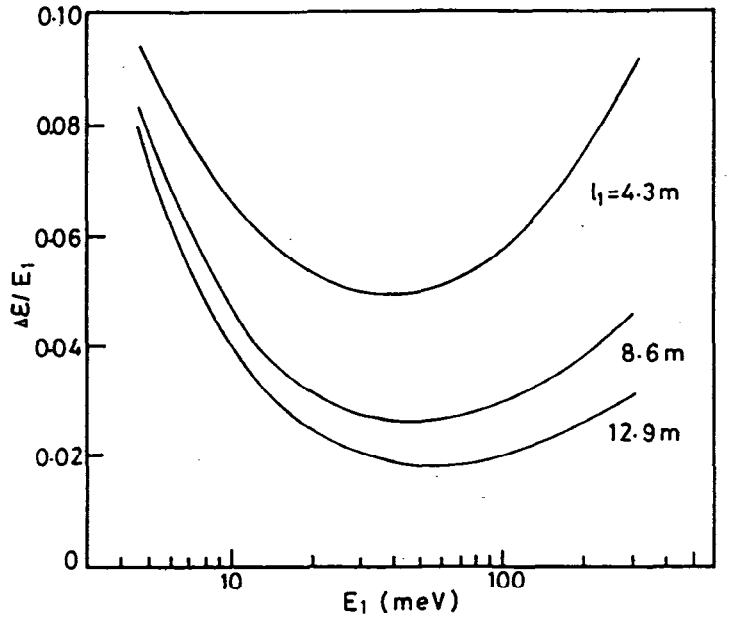

Fig.4. Incident energy dependence of overall resolution of LAM-D calculated for l_1=4.3, 8.6 and 12.9 m. Other parameters are the same as in Fig.2.

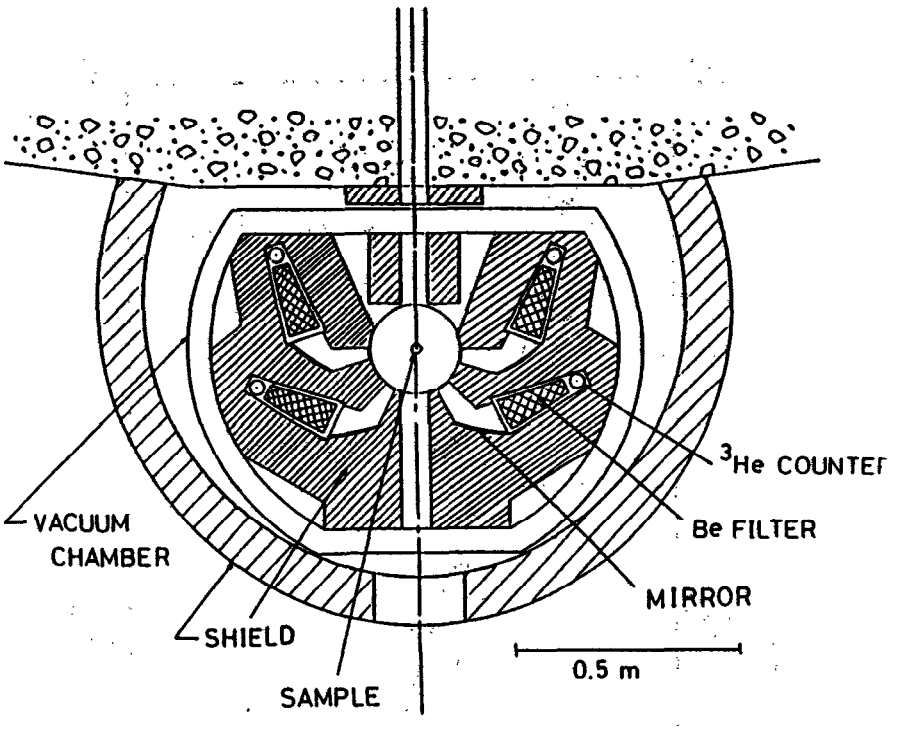

Fig.5. The inelastic spectrometer LAM-D.

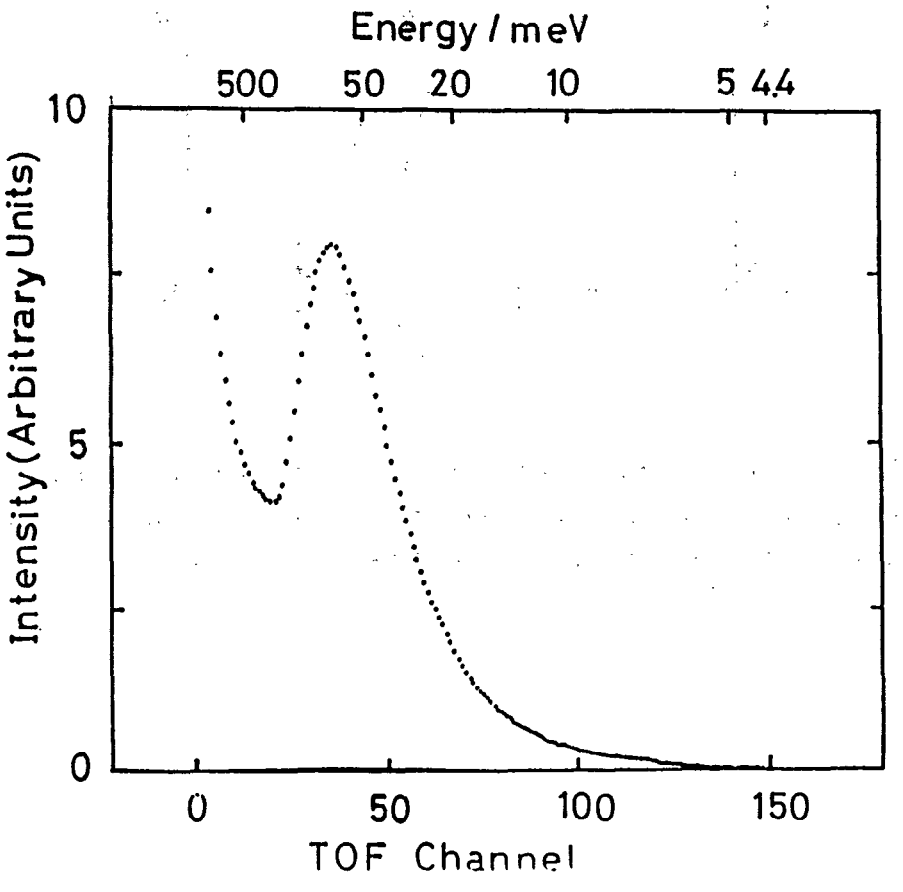

Fig.6. Energy distribution of neutrons from KENS thermal neutron source; H_2O at room temperature.

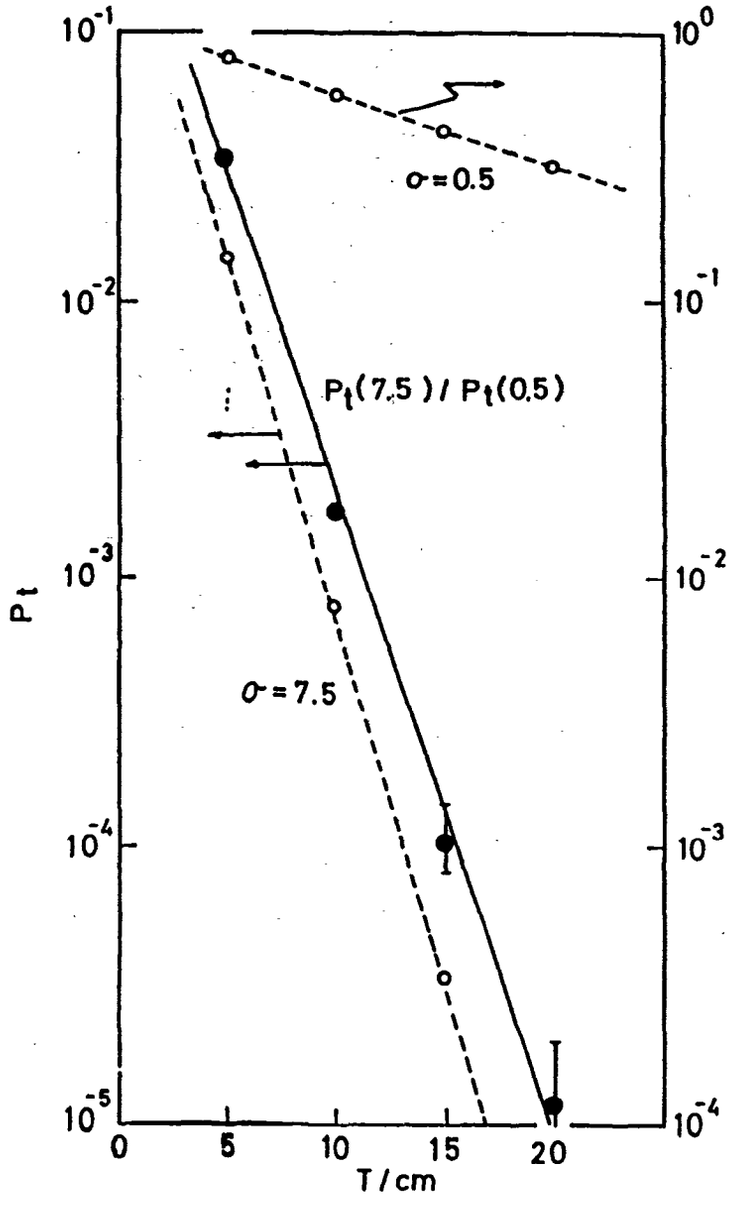

Fig.7. Results of the Monte Carlo simulation for neutron transmission through Be-filter. Calculation was performed for neutrons below and above the Be cutoff wavelength. The corresponding atomic cross sections are 7.5 and 0.5 barns, respectively.

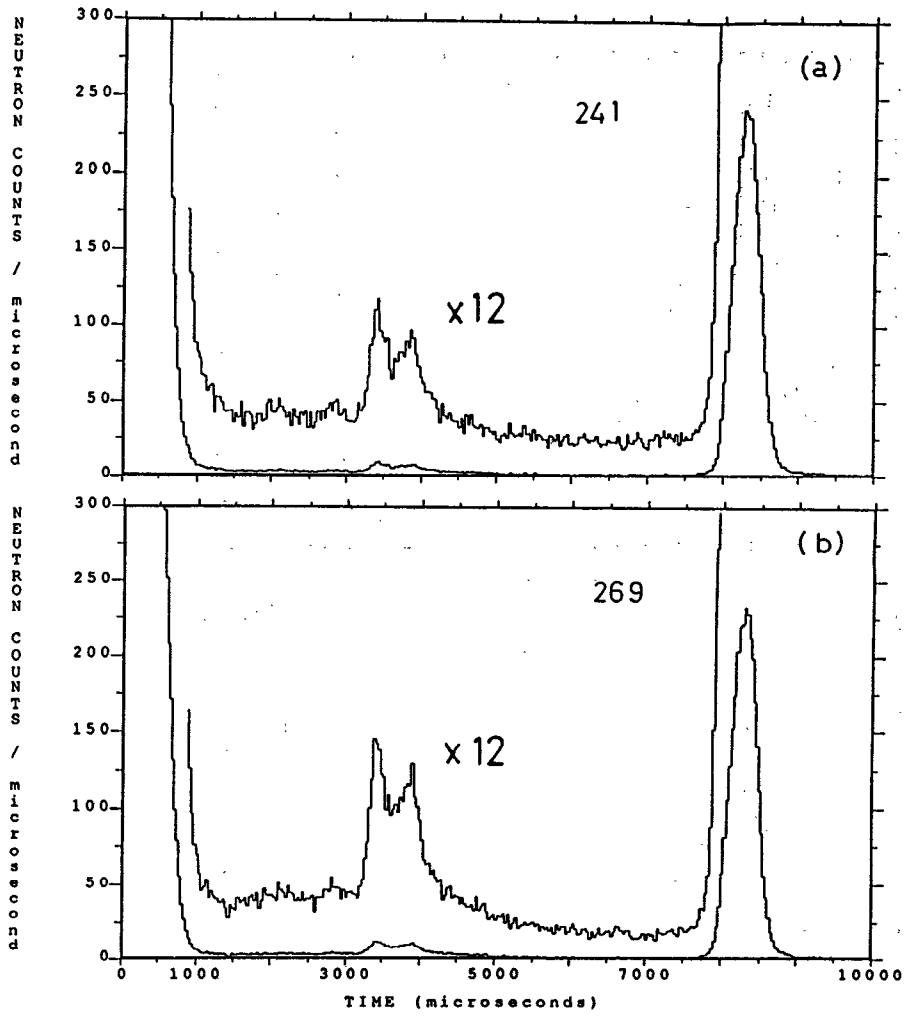

Fig.8. Spectra of vanadium measured by LAM-D. The thickness of the vanadium sample is 2 mm and the measuring time is 8 hrs. (a): the scattering angle 35 deg, (b): 85 deg.

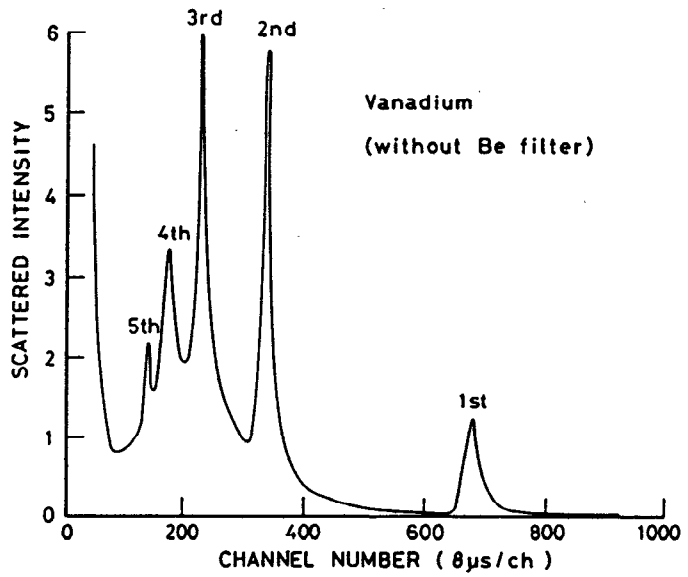

Fig.9. Spectra of vanadium measured LAM-D without the 15 cm Be-filter.

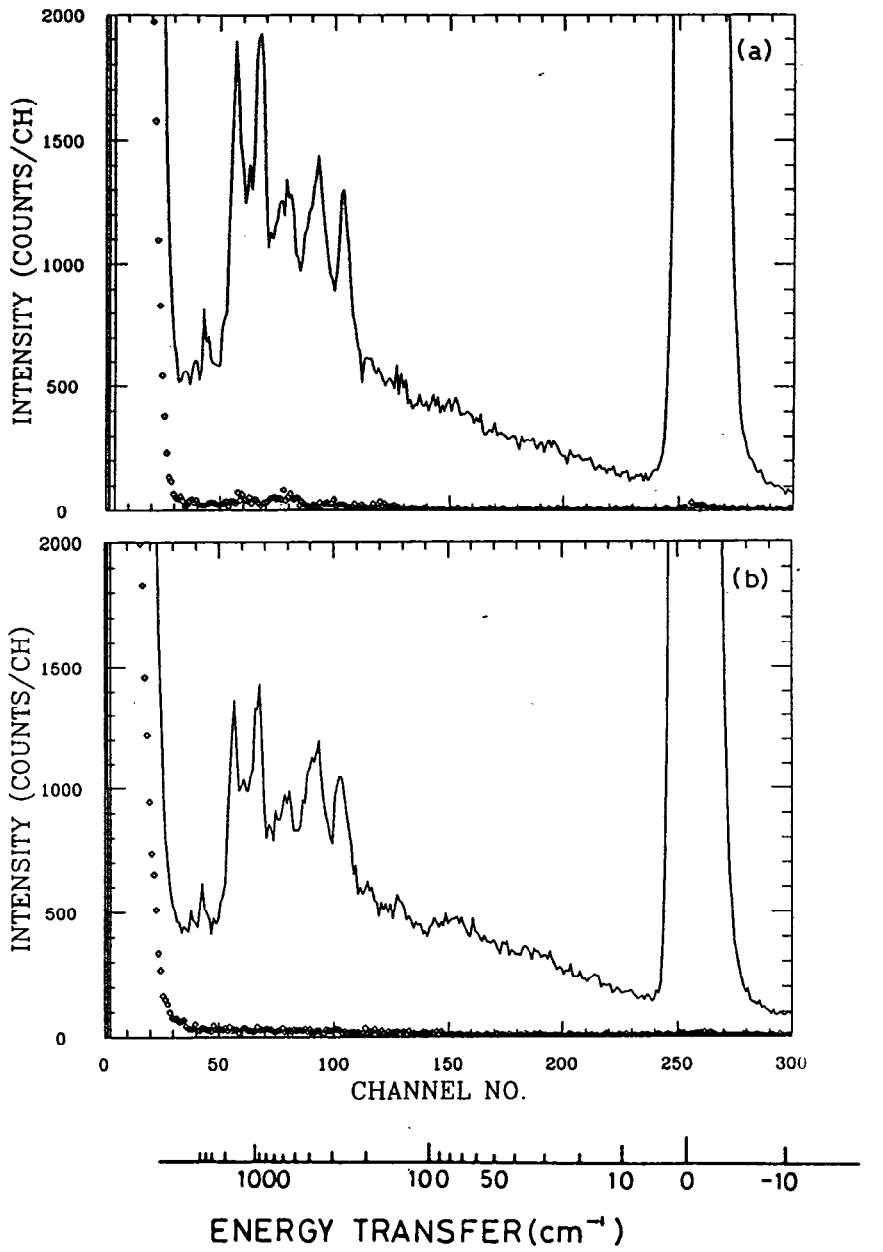

Fig.10. Inelastic scattering spectra of trans-1,4-polychloroprene at 10K and sample container which are normalized by monitor counts. (a): the scattering angle 35 deg, (b): 85 deg.