The use of chopper spectrometers for cold-to-epithermal neutron scattering at IPNS

C.-K. Loong, L. I. Donley, G. E. Ostrowski, R. Kleb,
J. P. Hammonds, L. Soderholm, and S. Takahashi*
Argonne National Laboratory, Argonne, Illinois 60439-4814, U. S. A.

Abstract

A multi-detector chopper spectrometer enables measurements of the scattering function S(Q,E) to be made over a wide range of momentum and energy transfer (Q,E). The application of pulsed-source chopper spectrometers for inelastic measurements at thermal and epithermal energies (50 meV < E < 1000 meV) is well known. Recently at IPNS, we have extended the energy-transfer region down to about 0.5 meV with a resolution of the order of 150 μeV. It is made possible by utilizing the cold-neutron incident spectrum of the 100 K methane moderator in conjunction with a dual beryllium-body rotor system. Neutron incident energies can be changed efficiently over the 4 to 1000 meV region while maintaining an undisturbed sample environment. We describe the operation of the IPNS chopper spectrometers (HRMECS and LRMECS), the instrumental resolution and the background-suppression performance. The capability of measuring inelastic features from 0.5 to 100 meV with an energy resolution of $\Delta E/E_0$ = 2.5% is demonstrated by experimental results of crystal-field excitation spectra of a high-T_c superconductor $ErBa_2Cu_3O_7$. Preliminary data of quasielastic scattering from a room-temperature molten salt $AlCl_3$-EMIC are presented.

I. Introduction

The dynamics of atomic and electronic systems of condensed matter is characterized by correlation functions over a large space-time domain. Neutron spectroscopy is capable of probing the dynamic response in a time scale of 10^{-8} to 10^{-14} sec, corresponding to an energy range of approximately 1 μeV to 1 eV. The coverage of spatial correlations, in terms of neutron momentum transfer $\hbar Q$, is determined by the scattering kinematics and the range of energies used. As far as experimental applications are concerned, the useful dynamic range of a spectrometer is affected by both the neutron source spectral distribution and the instrumental design. These factors have led to the design of a variety of instruments which are optimized individually for measurements in different regions of the (Q,E) space. As a result, experimenters are often required to perform measurements on the same samples using different spectrometers in order to collect the data needed to understand a system. For example, electronic transitions between states of transition-metal ion multiplets vary from a fraction of a milli-electron volt to many electron volts. Diffusional motion, atomic vibrations and deep-inelastic single-particle excitations within a molecular system have characteristic energies spanning decades in an energy scale. In view of the need, frequently encountered, to study the properties of a system on several different spectral and dynamic scales, using a single instrument would ease efforts in scheduling, experimental set-up, data normalization and interpretation. This paper describes the results in applying the IPNS chopper spectrometers in a range of energies widely extended from what was originally conceived.

A pulsed-source multi-detector chopper spectrometer is a versatile instrument for two main reasons: 1) large fluxes of cold-to-epithermal neutrons are available from a pulsed source equipped with cold moderators; and 2) chopper (rotor) systems are an effective means to select neutron incident energies from a few meV to 1 eV. The applications of pulsed-source chopper spectrometers for inelastic measurements at thermal and epithermal neutron energies were recognized in the beginning of pulsed-source development.[1] Their routine usage in the cold-neutron regime, on the other hand, has been realized only recently. We will describe the operation of a dual chopper system, which enables cold-to-epithermal neutron scattering on the IPNS chopper spectrometers HRMECS and LRMECS, and present some recent scientific results of quasielastic, near elastic and inelastic scattering.

II. Experimental Details

A chopper rotates at an angular frequency that is a multiple of the source repetition rate, i.e., from 30 Hz (fundamental) to 270 Hz (the 9th harmonic) at IPNS. The chopper aperture lines up with the source several times during the 33⅓ msec between successive bursts, but the curved multi-slits of the chopper window select a narrow energy-band of neutrons only at one opening. The neutron-absorbing slats are effective for energies up to a few eV. Higher-energy (fast, in the KeV range) neutrons at the time of the prompt burst are removed by scattering from the beryllium body of the rotor. Conceptually only one chopper is sufficient for the production of a monochromatic neutron beam. Unfortunately, the source also produces a small component of time-independent fast delayed neutrons that can pass through the chopper slit package at every opening. These fast neutrons scattered by substances intercepting the beam (container windows, samples, etc.), give rise to a background at the detectors with a time structure characteristic of the chopper's secondary openings, as shown in Fig. 1a. Moreover, the background level depends on the amounts of materials exposed to the beam, thus it cannot be corrected by using empty-cell runs.

1. t_0-choppers

A two-chopper system is called for to remedy the background problems. In this case a second, low-resolution chopper is placed upstream of the principle monochromating chopper to eliminate the secondary openings for delayed neutrons and to scatter prompt fast neutrons from the beam at a location farther from the sample than the principle chopper. The resulting background level, in a test run in 1987 as shown in Fig. 1b, improves significantly even though at that time the upstream chopper was not designed for optimal delayed neutron suppression. In 1988 the first background suppression chopper (coined "t_0-chopper", as opposed to the high-resolution energy selecting "E_0-chopper") was fabricated and operated on the PHENICS chopper spectrometer at IPNS. The t_0-chopper has a slightly larger beryllium body than that of the E_0-chopper but has no slit package, (see Fig. 2). Both the t_0- and the E_0-choppers are mounted in identical housings and controlled by the same drive system. The t_0-chopper serves to remove the delayed and the prompt-pulse fast neutrons. Currently, all three chopper spectrometers at IPNS are equipped with similar t_0-choppers.

2. E_0-choppers

A unique feature of IPNS E_0-choppers is the large (up to 7.5 X 10 cm) aperture of the slit packages and the rather massive beryllium body, which provide effective stopping power for neutron removal. The disadvantage of such a design is the large energy required for high-speed rotation. The current drive system using mechanical bearings constrains the maximum speed to 270 Hz. An important criterion is to match the chopper opening time with the neutron pulse width so as to maximize the intensity without sacrificing the resolution. If a chopper is to be maintained at the maximum speed (always desirable in principle), it requires different slit packages for different energy selection, i.e., larger slit radii of curvature and tighter slits at higher energies. Six E_0-choppers have been built for the three chopper spectrometers at IPNS.[2] The energy resolution $\Delta E/E_0$ varies from about 4% at the elastic position for HRMECS and PHENICS (7% for LRMECS) to about 2% near the end of the neutron-energy-loss spectrum for HRMECS and PHENICS (4% for LRMECS). Better resolution, especially for $E_0 > 300$ meV, can be achieved by running with higher chopper speeds but this would require a new drive-control system.

3. Variable-speed dual chopper operation

Two major factors come into play for low energy ($E < 50$ meV) operations: 1) The pulse widths are broader so it allows a chopper window assembled with wider slits (and less curvature); and 2) The lower neutron speeds require much extended background-free time fields for energy analysis (e. g., over 20,000 μs for a 4-meV run on HRMECS). If a chopper maintains the same speed, say 270 Hz, a change of the neutron energy requires switching E_0-chopper during the experiment. More importantly, it becomes difficult to eliminate all the simultaneous openings of both the t_0- and E_0-choppers for fast delayed neutrons over such large time intervals.

The conditions for low-energy operation can be accommodated easily if the choppers can be run and controlled at lower speeds. An E_0-chopper that is optimized for high energy at 270 Hz operation becomes an efficient chopper for lower energies when its speed is reduced to lower harmonics of 30 Hz. At a lower angular speed, the sweeping time of the chopper slits across the beam increases and the curved slits transmit neutrons having a lower energy. Moreover, a lower chopper frequency reduces the number of secondary openings, which makes background control manageable over the larger time field (see Fig. 3). Changing chopper speeds takes a fraction of an hour as compared to a several hour job for switching choppers. Consequently, a variable-speed dual rotor system enables efficient operations between 4 to 1000 meV without a significant loss of intensity or resolution. The sample environment is not disturbed throughout measurements using cold-to-epithermal neutrons.

In 1991 the chopper controller circuitry was modified to allow chopper rotation at any harmonics between 30-270 Hz.[3] We find that the IPNS 100 K methane moderators provide usable fluxes of neutrons for effectual experiments from 4 meV to 2 eV.[4] We show in Fig. 3 the neutron "time schedules" for 110 and 4 meV runs using the same t_0- and E_0-choppers on HRMECS. Table I lists examples of chopper speeds usable for HRMECS and LRMECS experiments over the 4 to 400 meV energy region.

Table I. Examples of chopper angular frequencies to be used for HRMECS and LRMECS experiments with incident energies ranging from 4 to 400 meV. The same E_0-chopper (the 250-3 rotor for HRMECS and the 160-2 rotor for LRMECS) is used throughout for each spectrometer.

Incident energy	HRMECS		LRMECS	
(meV)	t_0-chopper (Hz)	E_0-chopper (Hz)	t_0-chopper (Hz)	E_0-chopper (Hz)
4	90	30	60	30
8	30	60	60	60
20	30	90	60	90
40	60	90	90	120
60	60	150	120	180
80	60	150	120	180
110	90	210	120	210
140	90	210	150	240
170	90	240	150	270
200	90	240	150	270
250	90	270	150	270
400	90	270	150	270

III. Results and Discussion

We present some recent HRMECS and LRMECS experimental results to illustrate the merits of performing measurements using cold-to-epithermal neutrons on the same spectrometer. Fig. 4 show the observed excitation spectra of a magnetic superconductor $ErBa_2Cu_3O_7$.[5] Four incident energies, 4, 8, 20 and 110 meV, were used to resolve transitions between the crystal-field states of the Er^{3+} ionic ground multiplet which spans an 80-meV interval with uneven splittings (see Fig. 5). It is essential to perform experiments with a wide range of incident energies at various temperatures in order to confirm the energy level structure. Furthermore, since the data were taken from the same spectrometer, the observed intensities were normalized in a straightforward manner, and the whole data set can be compared quantitatively with results from model calculation.[5]

Fig. 6 displays the quasielastic spectra for a room-temperature molten salt $AlCl_3$-1-ethyl-3-methyl imidazolium chloride (EMIC) measured by LRMECS with an incident energy of 4 meV.[6] The quasielastic scattering, measured as a function of Q from 0.2 to 2 $Å^{-1}$ at 25 ° C, provides information regarding to the diffusional motion of the molecular species in the liquid phase. The inelastic spectra[6] (not shown), measured with E_0 of 60, 160 and 400 meV at 25 K and room temperature, reveal features corresponding to collective excitations and internal molecular vibrations which afford a quantitative comparison with results of *ab initio* calculations.[7]

In summary, we describe the application of a dual chopper system on HRMECS and LRMECS for energy selections from the cold-to-epithermal neutron spectrum provided by the IPNS 100 K methane moderators. We also present some results of recent scientific studies which have taken advantage of this improved operation of the chopper spectrometers.

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* On leave from Nisshin Steel Co. Ltd., 7-1 Koya-shinmachi, Ichikawa, Chiba 272, Japan.

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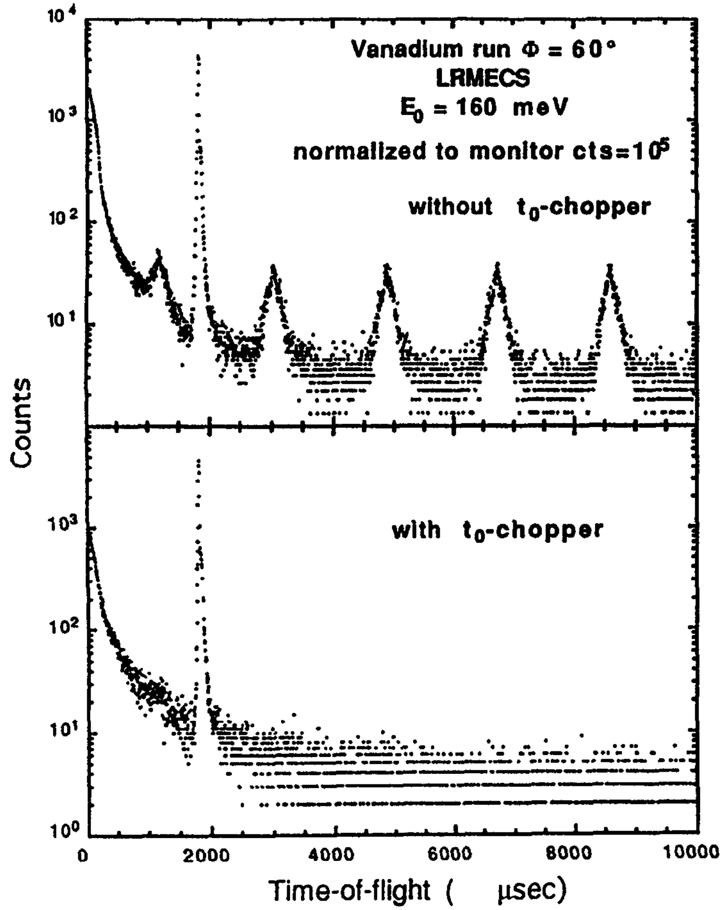

Figure 1. Observed detector spectra of a vanadium standard run (a) without and (b) with a background suppression t_0-chopper.

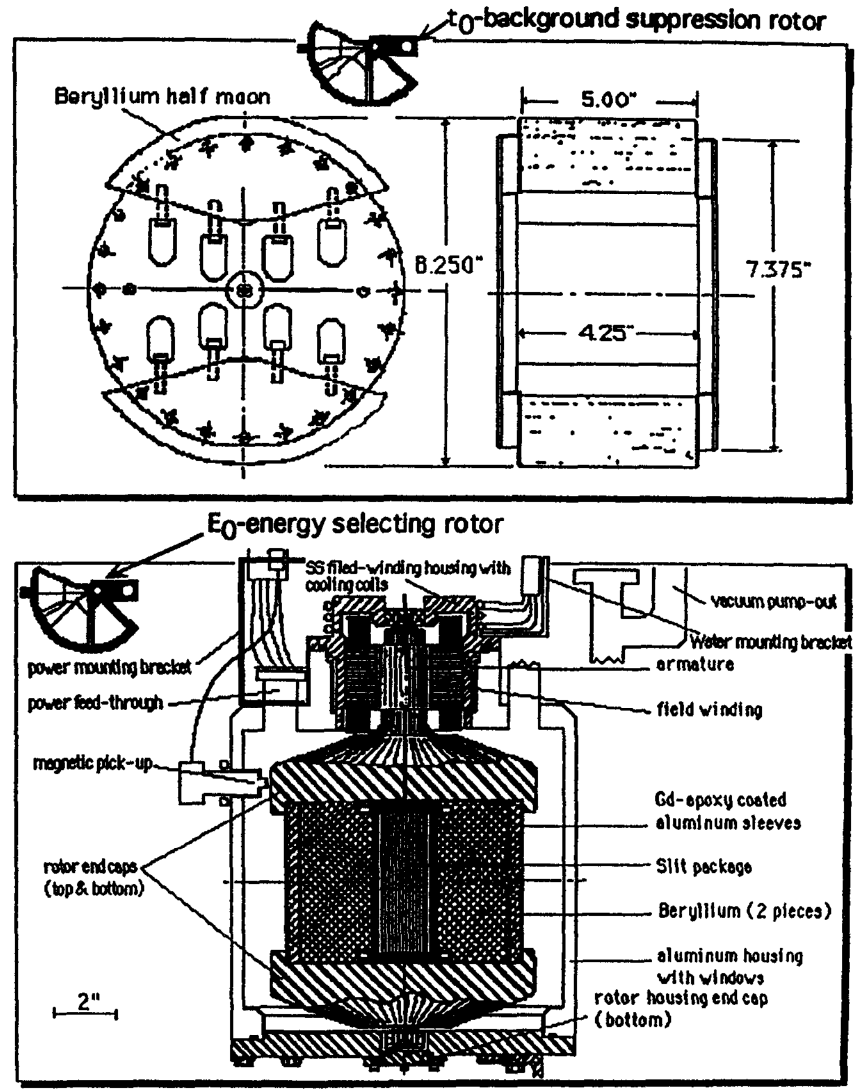

Figure 2. The IPNS t_0-background suppression chopper and the E_0-energy selecting chopper (with housing).

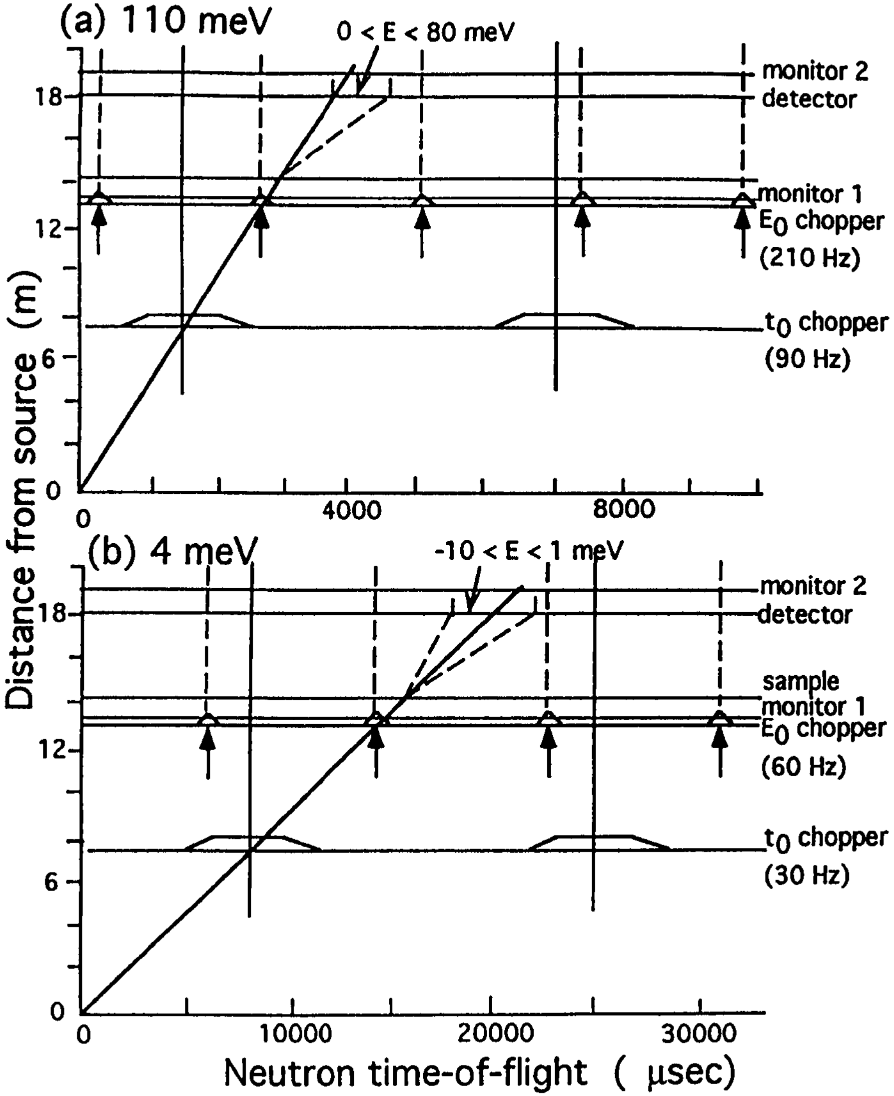

Figure 3. Neutron flight distance versus time for (a) 110 meV and (b) 4 meV operation. The E_0-chopper, optimized for 250 meV at 270 Hz, can be effectively used for 110 and 4 meV at reduced speeds.

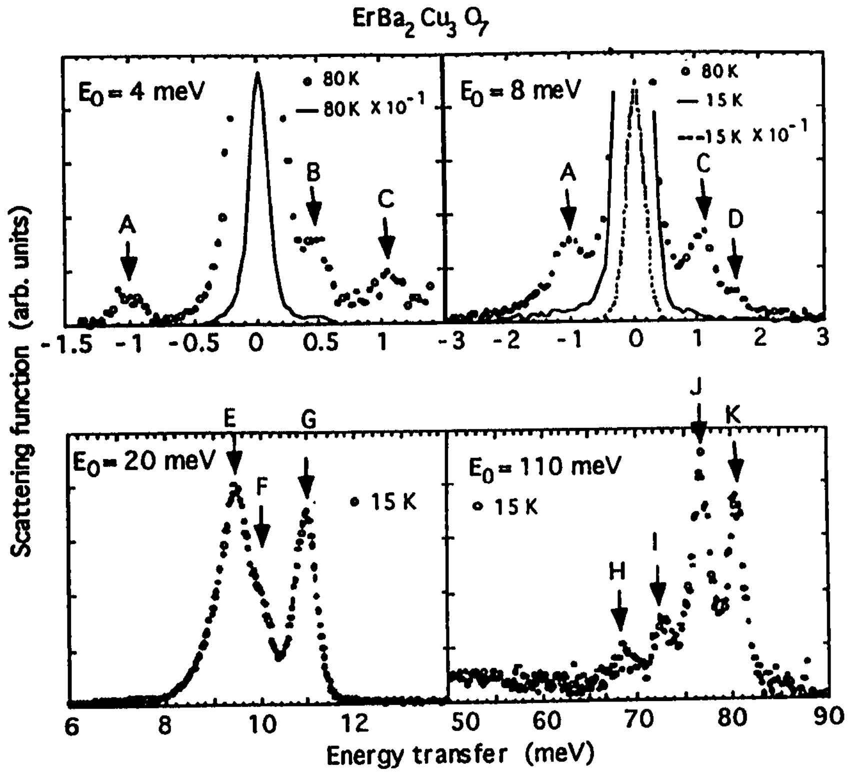

Figure 4. The observed excitation spectra of magnetic superconductor $ErBa_2Cu_3O_7$. The peaks labeled correspond to crystal-field transitions between the energy levels of the Er^{3+} ionic ground multiplets $^4I_{15/2}$, as shown in Fig. 5.

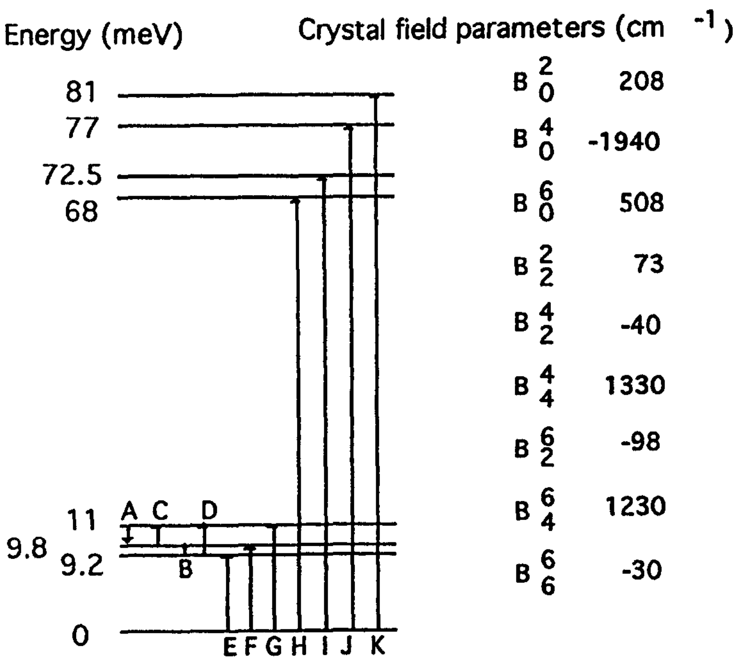

Figure 5. A schematic diagram of the crystal-field levels for the ground multiplet $^4I_{15/2}$ of Er^{3+} ions in $ErBa_2Cu_3O_7$. The transitions labeled correspond to the observed peaks shown in Fig. 4, and the B_q^k are the crystal-field parameters derived from the neutron data.

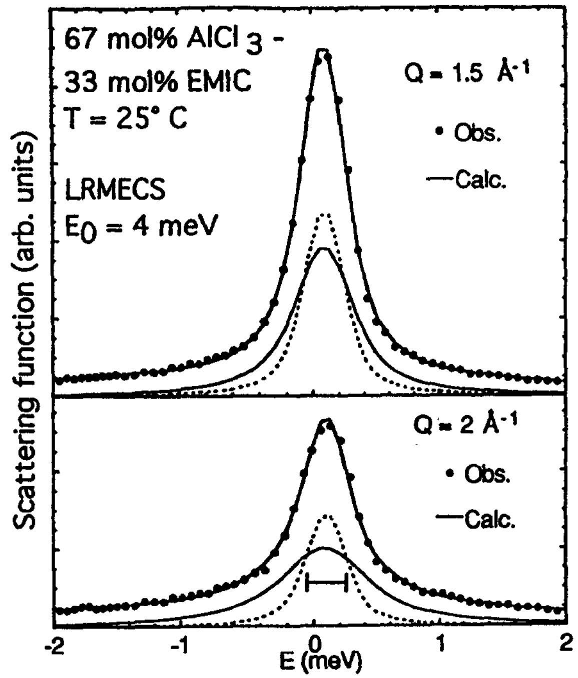

Figure 6. The quasielastic spectra for 67 mol% $AlCl_3$ - 54 mol% EMIC obtained using LRMECS with an incident neutron energy of 4 meV. The data were analyzed by a composition of a sloppy background and two Lorentzian functions.[6] The energy resolution (FWHM) is indicated by the horizontal bar in the lower panel.