Options for disordered materials diffraction at the ESS – a discussion document

Background

Over the past several decades, Europe has led the World in the provision of diffraction facilities for liquids and non-crystalline solids. Diffractometers appropriate to this work include D4 and D20 at ILL, 7C2 at Saclay, and SANDALS at ISIS. In addition a state-of-the-art diffractometer, GEM, has recently been installed at ISIS, and there are somewhat lower flux facilities such as SLAD at Studsvik. Between them these instruments have studied a very broad range of disordered materials science which has been widely disseminated in the scientific literature, including prestigious journals such as *Nature*, *Science*, and *Physical Review Letters*.

The theme of $S(Q)$ measurements actually covers a rather broad front from the measurement of the radial distribution function in a simple fluid, like argon or krypton, through the study of a range of increasingly complex fluids and liquid mixtures, including supercritical fluids, molten salts, liquid metals, liquid semiconductors and ceramics, liquid mixtures and solutions, molten and amorphous polymers, heterogeneous systems (e.g. liquids absorbed in solids), amorphous and glassy solids. Total scattering studies of crystalline materials could also be included in this list. Because of this diverse range of materials, the requirements on the diffractometer are equally diverse. High Q (≥ 60Å^{-1}) with high resolution are essential for amorphous materials where the structure is often sensitive to subtle changes in the near-neighbour distances between atoms. On the other hand low or very low Q (~ 0.01Å^{-1}) values are increasingly needed to probe the longer range order present in many technologically important complex fluids. What has not been achieved to date is the combination of very high and very low Q capability in a single diffractometer, and it is this possibility which will set an instrument built at the ESS apart from anything that can be envisaged at present day facilities.

Requirements for a Disordered Materials Diffractometer at the ESS

Based on the experience gained at existing facilities it is now possible to state clearly what features any new diffractometer(s) in this field will require:-

1. A very wide Q range:-(0.01Å$^{-1} < Q < 60$ Å^{-1})

2. Good Q resolution (depending on the Q value):-(5-10% $\geq \Delta Q/Q \geq$ 0.5%(?))

3. Good count-rate, at or above 100 cts/s/0.05Å^{-1}/per cm^3 vanadium over the whole Q-range.

4. Acceptable recoil corrections.

5. Low backgrounds.

6. Very stable detectors (<0.1% drift over 24hrs). This condition applies equally to the beam monitors. Assuming stable monitors and detectors are available small drifts in the incident spectrum are acceptable at the 1-2% level.

7. Rapid means of changing samples and sample conditions.

8. Rapid data accumulation (fast "begins" and "ends") plus rapid data assessment/correction/reduction. It was felt that the rapid assessment of data is essential

for any future diffractometer since how to proceed with any given experiment often depends on appreciating how a particular sample appears from the diffraction pattern.

9. Sophisticated data interpretation tools, so that the structural consequences of any diffraction data can be assessed promptly as soon as they are accumulated.

Consequences for a Disordered Materials Diffractometer at the ESS

1. ***Q*-range**: The very wide *Q* range can only be achieved with a combination of a range of scattering angles with full exploitation of the neutron energy range available at the ESS.

2. **Large *Q*:** This is generally only achievable with high resolution in the backscattering direction. However lower resolution is achievable at large *Q* in the forward direction if the full moderator spectrum at high energies is available. This precludes the possibility of using a T0 chopper to block the fast neutrons from the experiment.

3. **Low *Q***: Requires longer flight path to reduce beam divergence and so keep resolution at low *Q* acceptable.

4. **Resolution**: Neutron pulse width should ideally be matched to geometric resolution. This can be hard to achieve in an instrument which has both wide angle and low angle detectors. The notion of (almost) constant resolution detectors has proved useful at ISIS when banks of detectors are to be merged together. It is particularly important from the point of view of data interpretation of structures with sharp peaks that the resolution function is correctly included in the data interpretation process.

5. **Count rate**: This can only be achieved by means of large solid angle detector arrays. If the source is short-pulsed the detectors do not need to be moved, which means the solid angle can be as large as cost and engineering constraints allow.

6. **Recoil corrections**: Keeping these small means keeping the scattering angle below about 50° and neutron energies above the thermal energy of the sample. In some special cases (large scale structures for example or Bragg scattering) the latter condition can be relaxed somewhat.

7. **Low backgrounds**. Effective collimation is needed on both the incident and scattered beams.

8. **Detectors and data acquisition electronics**: As well as being STABLE these must also be FAST, i.e. low deadtimes. (On ISIS at ~160kW, the DAE_I system falls over when the time averaged countrate exceeds about 1MHz.) With ESS running at roughly 20 times this countrate, there will be serious deadtime problems unless much fast electronics can be incorporated. **It is particularly important to set up an electronics regime whose deadtime is quantifiable, since the variation in deadtime from different scattering samples will have serious deleterious effects on the overall reliability of the data if the deadtime cannot be quantified.**

Comparative performance of two generic disordered materials diffractometers.

Based on the spectral parameters produced by Feri Mezei (4-12-2000), two generic diffractometers were analysed with a simple Monte Carlo routine to give comparative estimates of the likely resolution and count rates on the ESS. These are compared to estimates of what is currently available on the SANDALS diffractometer at ISIS, based on the published spectral parameters for the ISIS methane moderator. It should be stressed that the actual count rates (in particular) and resolution will depend on a number of factors which will only be known once experience with the actual target/moderator configuration becomes available.

In both cases it was assumed the disordered materials diffractometer (DMD) would consist of a large solid angle of detectors spanning the scattering angle range 0° - 40°, plus a smaller bank of detectors in the backscattering direction (150°). The detectors at low angles were arranged on the surface of a cylinder of radius 0.8m from the beam axis, for scattering angles above ~11°. For lower angles than this the secondary flight path was assumed to be ~6m from the sample. Individual detector elements were 200mm high by 10mm wide (facing the sample) by 20mm deep, except at the very lowest angles, where they were assumed to be either 50mm or 100mm in height. The sample was a 1cm^3 of vanadium. It was assumed the incident beam would be circular in cross section, with the viewing area of the moderator a disk of diameter ~90mm. Absorbing apertures were placed 1.7m from the moderator and 0.3m from the sample to simulate the effect of the collimator on instrument performance.

For DMD-I, the incident flight path was set to 11m, the ambient water moderator was viewed, and it was assumed the instrument would be on the 50Hz target. Figure 1 shows the estimated resolution for the three types of moderator (decoupled/poisoned, bottom; decoupled/unpoisoned, middle; coupled/unpoisoned, top). Figure 2 shows the estimated count rate (or “C-number”) for each moderator, and the circles show the comparative performance of the current SANDALS, estimated in the same way.

For DMD-II, the incident flight path was set to 25m, the liquid hydrogen moderator was viewed, and the instrument was assumed to be on the 10Hz target. Figs. 3 and 4 are the sequel to Figs. 1 and 2 for this configuration.

Discussion

It is clear that based on this rather simple comparison, a disordered materials diffractometer on ESS will achieve count rates up to 20× what is available at existing facilities, with an equivalent resolution. Such an instrument would also have a wider *Q* range with a given count rate than any existing diffractometer.

The 11m option DMD-I gives an exceedingly high count rate gain over existing machines, but there are only minor gains at low *Q* because the water moderator available cuts out too soon at low neutron energies. This could be in principle be won back by using a hydrogen moderator, but then there will be significant frame overlap which would necessitate the use of frame overlap choppers.

The 25m option DMD-II apparently gives very little count-rate advantage over existing machines, but what these graphs fail to show is that the **minimum *Q*** that would be available on this machine would be at least a factor of 2 smaller than for DMD-I. This is because the longer flight path permits a narrower beam divergence, which in turn allows smaller scattering angles to be accessed. In addition by running on a 10Hz target the full range of neutron energies can be utilised.

The current view is that **neither** of these arrangements is optimal. DMD-I shows good count rate gains at large *Q*, but will not extend the minimum *Q* value significantly. DMD-II will achieve the required minimum *Q* but at substantial expense in count-rate. In fact on this flight path (~30m overall), the 10Hz repetition rate is too slow, and some of the flux loss could be recouped by running at say 25Hz instead. Use of frame choppers to chop alternate pulses on the 50Hz target in order to run at 25Hz could be explored although there is no experience of this on existing disordered materials diffractometers. Frame choppers will never completely attenuate the power pulse, leading to scattered neutron spectrum with a pronounced spike in it. By exploiting the wide angle range of the DMD it might be possible to eliminate this in the final diffraction pattern, but there is no experience so far in attempting to do this.

The ideal would be to have a liquid methane type moderator on a 50Hz target, or else a liquid hydrogen moderator running at 25Hz.

As far as resolution is concerned it appears that the preferred moderator is decoupled and poisoned, although the unpoisoned moderator will deliver excellent count rates at slightly poorer resolution. The coupled moderators are definitely ruled out for $S(Q)$ work.

Long pulse target

The 2ms pulse width on the long-pulse target is too long to be used for $S(Q)$ diffraction directly. Therefore it would be necessary to monochromate the beam, much in the same way as is done currently at a reactor source. Much of the gain from using a pulsed source is then thrown away with this option. The combined goals of obtaining a wider Q range with increased count rate could only be achieved in this case by repeating each diffraction measurement at a sequence of incident energies to get the fullest range possible. Given the often delicate nature of many of the new samples likely to be studied (examples of this are the recently executed levitator experiments) this is not always a practical method of operation. In addition the goal of reducing recoil corrections by going to low scattering angles and high neutron energies is not so easy to achieve in this case. Therefore for the purpose of this study, the exploitation of the long pulse target was ruled out as a possibility.

Summary

Although substantial gains compared to existing sources are achievable at ESS, neither of the options of 50Hz operation on a water moderator or 10Hz operation on a hydrogen moderator are ideal for the next generation of disordered materials diffractometers. To make real progress this field needs very stable detectors and electronics, low backgrounds, and a very wide range of Q with appropriate resolution. Existing diffractometers on reactor sources provide good count rates and stability but do not permit very low or very high Q measurements. Diffractometers on pulsed neutron sources achieve a much better Q range with adequate count rates, but often have not achieved a detector stability comparable with the reactor instrument. (The true performance of GEM has yet to be established in this regard.)

The ideal disordered materials diffractometer will need a liquid methane type moderator at 50Hz or a liquid hydrogen moderator at 25Hz. The preferred option is for a poisoned moderator, but decoupled/unpoisoned could be considered as well. There is no use for coupled/unpoisoned moderators for this work. The long pulse target could be used in reactor mode, with a monochromator, but there is little gain in either count rate, resolution or Q range in doing this.

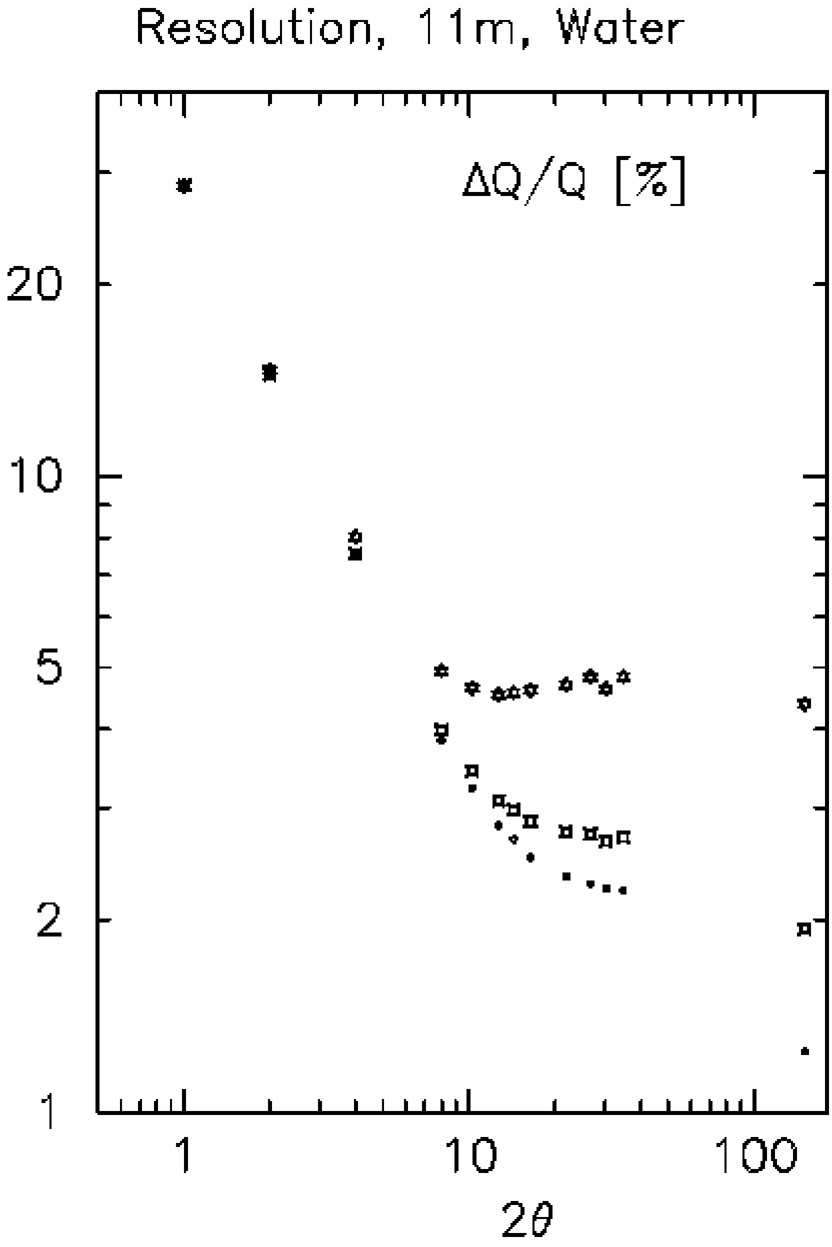

Figure 1. Resolution of DMD-1, 11m incident flight path. The dots correspond to decoupled/poisoned moderator, the squares to the decoupled/unpoisoned moderator, and the stars to the coupled/unpoisoned moderator.

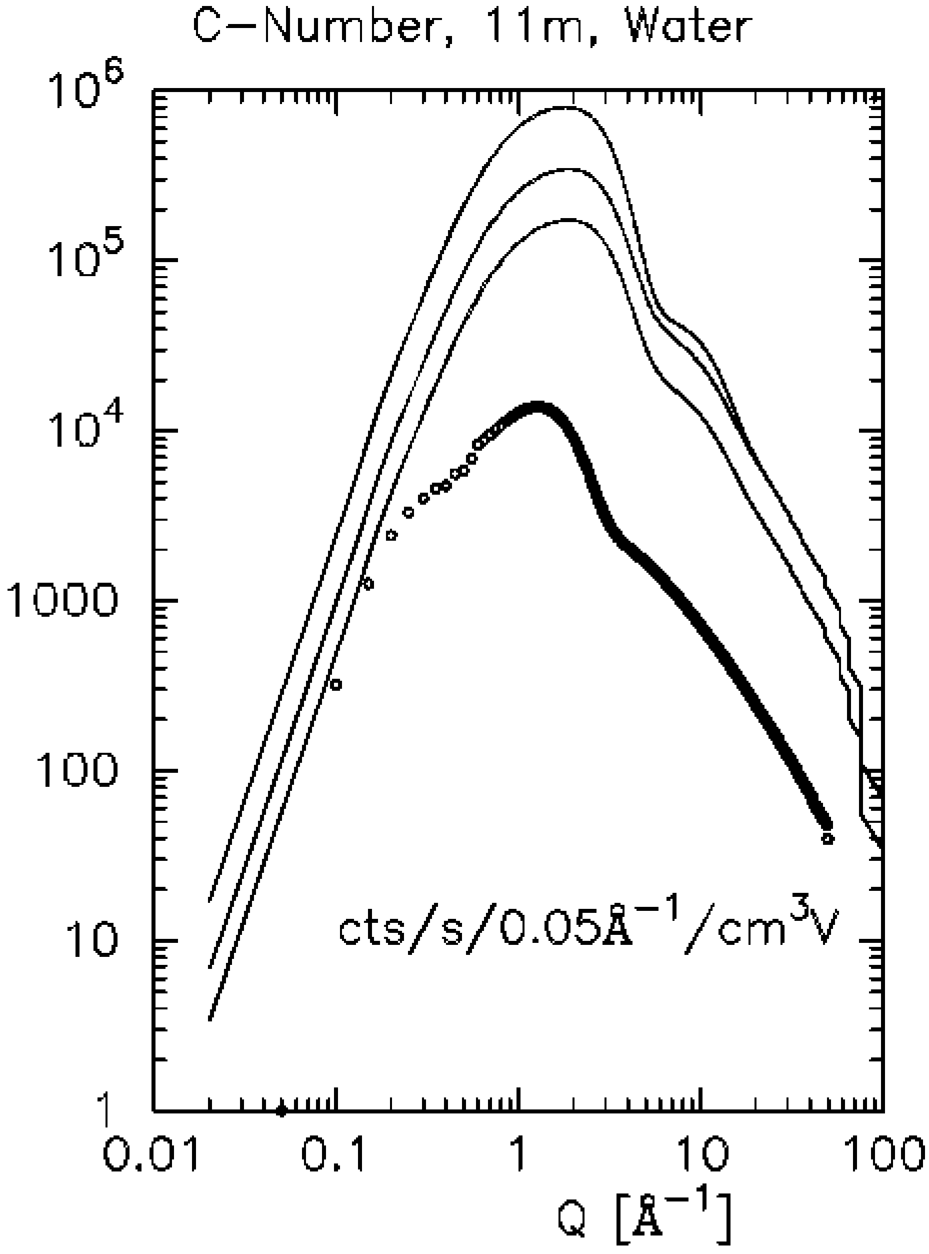

Figure 2. Count rate number for DMD-I as a function of wave vector transfer. The circles correspond to the current SANDALS estimated using the same procedure as described here. The top line is for the coupled/unpoisoned moderator, and the bottom line is for the decoupled/poisoned moderator.

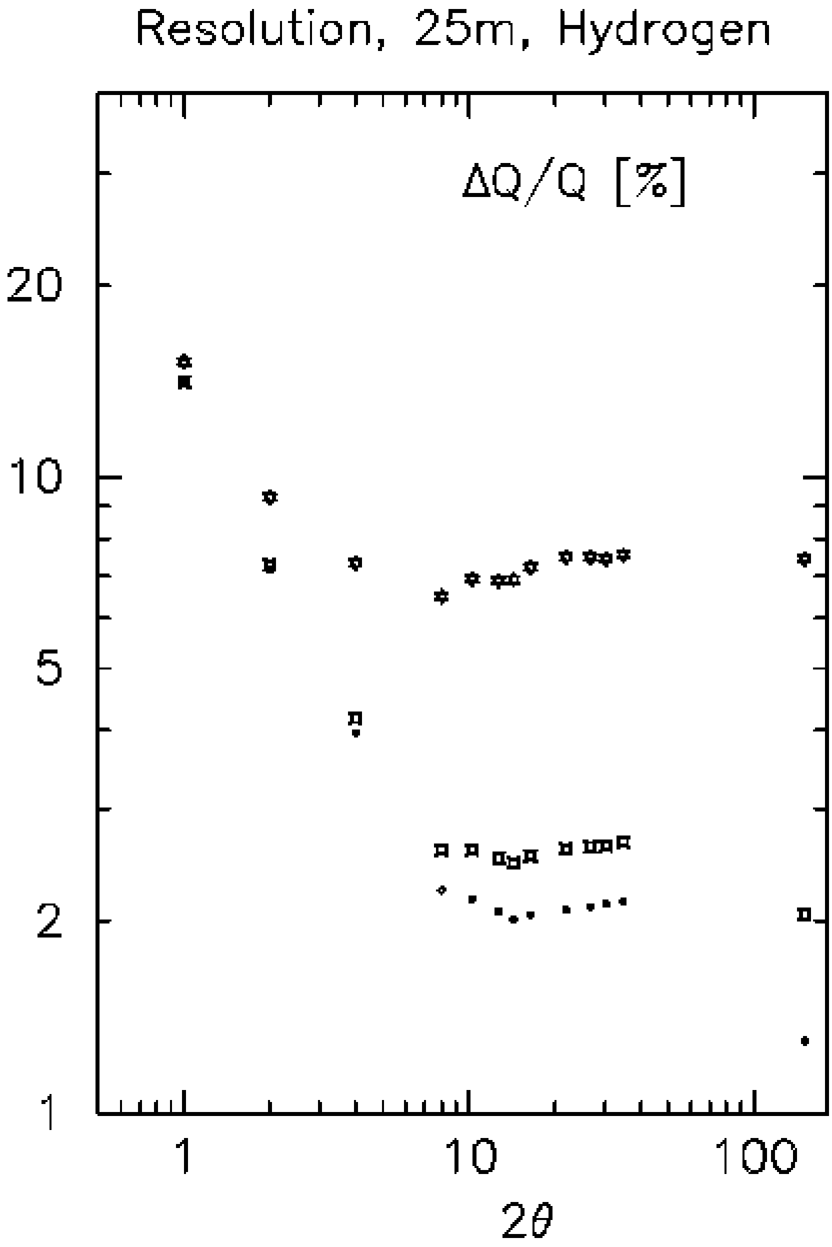

Figure 3. Resolution for DMD-II. Symbols as for Fig. 1

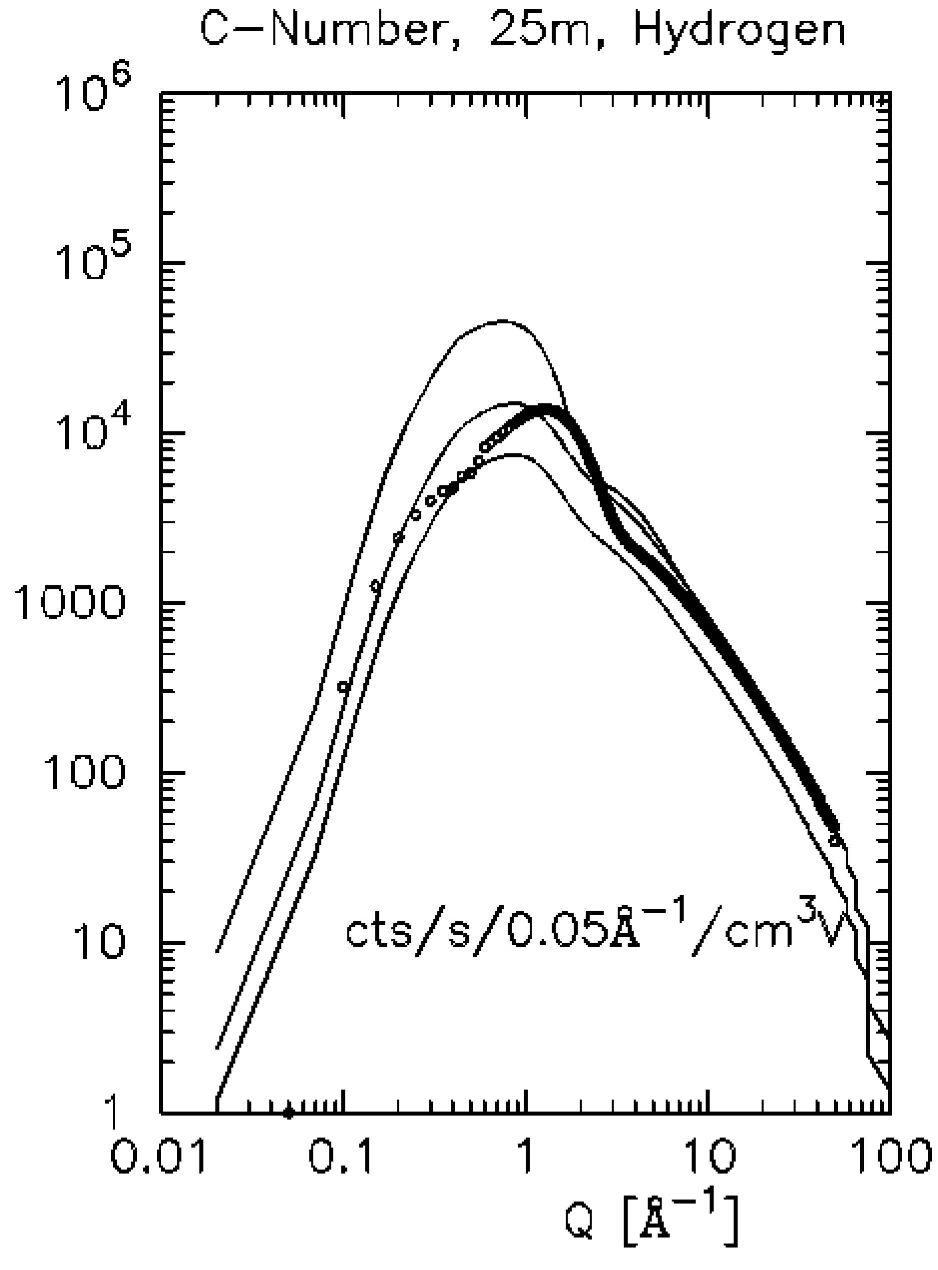

Figure 4. C-number for DMD-II.