

[TAS resolution]
(Chapter 6.3.2 in *Elements*)

Resolution

Cooper and Nathans (1967) have elegantly worked out the resolution function for the classic spectrometer (see also Shirane, 2002). The treatment of Popovici (1975) includes treatment of more general components, including curved monochromators and small samples. Here we outline the Cooper-Nathans procedure but refer the reader to the original papers for details. To a good approximation, the resolution function is a four-dimensional Gaussian form,

$$R(\varepsilon_o + \Delta\varepsilon, \bar{Q}_o + \Delta\bar{Q}) = R_o \exp\left(-\frac{1}{2} \sum_{k=1}^4 \sum_{l=1}^4 M_{kl} X_k X_l\right), \quad (6-TAS-1)$$

where $X_1 = \Delta Q_x$, $X_2 = \Delta Q_y$, $X_3 = \Delta Q_z$, and $X_4 = \Delta\varepsilon$, and

$$R_o = R(\varepsilon_o + \Delta\varepsilon, \bar{Q}_o + \Delta\bar{Q}) \Big|_{\Delta\varepsilon=0 \text{ and } \Delta\bar{Q}=0} = R(\varepsilon_o, \bar{Q}_o). \quad (6-TAS-2)$$

The prefactor R_o and the elements of the matrix M_{kl} , which is in general not diagonal, are involved functions of the incident wavelength λ_o , the set point ε_o, \bar{Q}_o and all the quantities that characterize the instrument: crystal plane spacings and mosaic widths, and collimator parameters (see Cooper and Nathans 1967, App. II; Shirane, 2002, App. 4). But the resolution function is a simple Gaussian for any straight-line path in ε, \bar{Q} space that passes through the set point ε_o, \bar{Q}_o . Moreover, the resolution in the \bar{Q} direction perpendicular to the scattering plane is usually irrelevant except insofar as it affects the measured intensity.

It is convenient to visualize the resolution function by considering the resolution function in terms of its constant-probability ellipsoid, say, its half-value locus, in ε, \bar{Q} space. These are called the resolution ellipsoids:

$$\exp\left(-\frac{1}{2} \sum_{k=1}^4 \sum_{l=1}^4 M_{kl} X_k X_l\right) = 0.5, \quad (6-TAS-3)$$

so that

$$\sum_{k=1}^4 \sum_{l=1}^4 M_{kl} X_k X_l = 2 \ln(2) = 1.386 \quad (6-TAS-4)$$

along the half-value locus.

Figure 6-TAS-2 illustrates a resolution ellipsoid for a setting that corresponds to $\Delta Q_z = 0$.

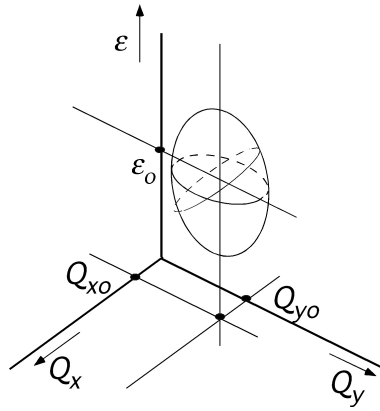


Figure 6-TAS-2. A representative resolution ellipsoid for a setting that corresponds to $\Delta Q_z = 0$.

Now is the hard part. When one is scanning to determine phonon dispersion relationships, it is desired to optimize the peak intensity and to minimize the width of the observed intensity distribution. The dispersion relation is a set of surfaces in \bar{Q} -space on which

$$\hbar\omega(\bar{Q}) = \hbar\omega_o, \quad (6\text{-TAS-5})$$

where $\omega(\bar{Q})$ is the *dispersion relation*, that is, the frequency of excitations as a function of wave vector \bar{Q} , and, for simplicity, the scattering function is

$$S(\bar{Q}, \varepsilon) = S_o \delta(\varepsilon - \hbar\omega(\bar{Q})) \quad (6\text{-TAS-6})$$

in the region of the measurement. In general there are a number of surfaces where such a relationship exists, several for each value of $\hbar\omega_o$, but if, for example, $\hbar\omega_o$ is very large, then no such surface exists. One exercise is to find values of $\hbar\omega_o$ that correspond to given \bar{Q} , a *constant- \bar{Q} -scan*. The experimenter sets the spectrometer for wave vector change \bar{Q}_o and varies the energy transfer ε until a peak appears. This establishes a point on the dispersion relation. (Usually there are several distinct branches of the dispersion relation.)

There are many ways to scan ε values with \bar{Q}_o fixed. High intensity indicates a large volume of \bar{Q} -space inside the resolution ellipsoid. But precise determination of the phonon frequency ω_o requires a narrow ellipsoid in the direction of the scan. The desired condition arises when the resolution ellipsoid is oblate, that is, pancake-like, with the thin axis parallel to the dispersion surface and the scan carried out in $\Delta\varepsilon, \Delta\bar{Q}$ space in the direction perpendicular to the dispersion surface. How can one do this? The dispersion surface is unknown, but the aim is to determine the dispersion function—the process is carried out iteratively, defining the surface crudely, with theoretical guidance as to the nature of the dispersion surface, then adjusting conditions to scan in the optimal direction.

For certain purposes, scanning \bar{Q} with constant ε , that is, *constant- ε scans*, is useful. This implies that k_1 and k_2 are fixed and \bar{Q} varies according to the directions of \bar{k}_1 and \bar{k}_2 in relation to the orientation of the sample crystal.

So much for preparing the spectrometer resolution ellipsoid. In phonon spectroscopy it is also necessary to arrange the \bar{Q} vector in relation to the direction of displacements $\bar{\xi}_i(\omega, \bar{Q})$ of atoms at position \bar{R}_i for normal mode ω so as to obtain the scattering intensity, which is proportional to the sum over atoms in the unit cell of $\left[b_i \bar{Q} \cdot \bar{\xi}_i(\omega, \bar{Q}) \exp(i\bar{Q} \cdot \bar{R}_i) \right]^2$ (see *Elements* Chap 10.4). In spectroscopy of magnetic excitations, the intensity in relation to excitations in the sample is more complicated, nevertheless requires consideration in setting up measurements (see *Elements* Chap. 4.5).

The procedures for calculating and adjusting the resolution function are now carried out by computer codes provided at each facility, based on the Cooper-Nathans analysis or the more general Popovici approach. At the outset of an experiment, the dispersion relations and eigenvectors are not known, although symmetry considerations may provide some guidance. It is a cut-and-try process, measuring to find roughly where the interesting features lie in ε, \bar{Q} space, then refining the setup, that is, the direction of the scans and the orientation of the resolution ellipsoids, with the help of the resolution codes, until a suitable instrument arrangement is established. Usually, a “W” configuration as shown in *Elements* Fig. 6.14 is found to be best.

During the course of measurement, interesting features frequently appear which are artifacts of the setup, not related to the physics of the sample and which must be cast out. Identifying these *spurious* tests the skills and intuition of researchers, whose duty it is to avoid being deceived and misleading others. Shirane 2002 devotes space to this process.

References

- M. J. Cooper and R. Nathans (1967). The resolution function in neutron diffractometry. I. The resolution function of a neutron diffractometer and its application to phonon measurements. *Acta Crystallogr.* **23**, 357-67 app 11.
- Popovici, M. (1975). On the resolution of triple-axis spectrometers, IV: The triple-axis spectrometer resolution function, special effects included. *Acta Crystallogr.* **A 31**, 507-13.
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