QENS models in STR_FIT

General remarks

All the QENS functions depend on Q (even the delta or the general Lorentzian may have a Q-dependence through the Debye-Waller factor). In order to facilitate the user input, the Q-dependence is not given in the parameter boxes (but in case of need, the value of Q can be used in the usual way, e.g. P1=2*(Q), as explained in the *Parameters Rules*). Instead, the current value of Q is passed to the function through the variable *Qcurrent1D* in the common *c_str_fitqens* and then it is handled by the function. **As a consequence, normally only S(Q,ω) spectra should be fitted using the models described here.**

No specific units are defined in the functions implemented, so thhe units mentioned here are given just as an example assuming that the input data will be S(Q,ω) spectra where Q is in Å^{-1} and ω in meV.

Most of the functions have at least one parameter that is Q-independent, so the 2D fit option should be used when fitting more than one spectrum. The notation in STR_FIT to say that a given parameter is Q-independent is to give its value between brackets, e.g. [1.5]. In the following, a physical parameter will be enclosed between brackets to show that the parameter should have the same value for all the spectra, e.g. [D] to indicate the self-diffusion coefficient.

All the standard models have the same initial four parameters (the indications given by the > and < signs are optional, but they are helpful to avoid that the fitting motor explores unphysical values):

P1: Scaling (intensity factor), >0 (intensity should always be positive).

P2: Position of the function (normally close to zero).

P3: Fraction (to use when using more than one function in order to define e.g. the fraction of atoms contributing to a particular function or the weight of that function), >0 <1 (it should be a fraction between 0 and 1). In many cases it should be independent of Q, so then it has to be given between [] and used with a 2D fit.

P4: ⟨u²⟩ to be employed to compute the Debye-Waller factor, >0. The Q-dependence of the DW term is taken into account by the function, so the initial value should be given between [] and the 2D fit employed.

1) Delta: Y = QENS_Delta (X, P1, P2, P3, P4)

Returns the intensity P1*P3*exp(-P4*Q^2/3) into a single energy channel corresponding to the position P2 or into two energy channels if P2 does not correspond to any of the X values passed to the function. In the latter case the intensity is divided between them accordingly to the distance between P2 and the two channels that enclose P2.

2) Lorenzian: Y = QENS_Lorentz (X, P1, P2, P3, P4, P5)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2/3)} \cdot \frac{1}{\pi} \cdot \frac{P_5}{(\omega - P_2)^2 + P_5^2}$$

General lorentzian of HWHM = *P5* and centered in *P2*.

3) Long range translational diffusion (random walk):

   Y = QENS_RandomWalkTranslation (X, P1, P2, P3, P4, P5)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2/3)} \cdot \frac{1}{\pi} \cdot \frac{P_5 Q^2}{(\omega - P_2)^2 + (P_5 Q^2)^2}$$

Brownian diffusion model with *P5* = [D] (self-diffusion constant in Å²·meV).

Definition

This model corresponds to a Brownian motion, where particles collide randomly between them. Between two collisions, one particle moves along a straight line. After a collision, it goes into a random direction, independent of the previous one. This requirement for 'memory loss' between two steps limits the minimum length and time that can be described by the model.

The corresponding scattering law is:

$$S(Q, \omega) = \frac{1}{\pi} \cdot \frac{DQ^2}{\omega^2 + (DQ^2)^2}$$

where *D* is the self-diffusion coefficient.

Example
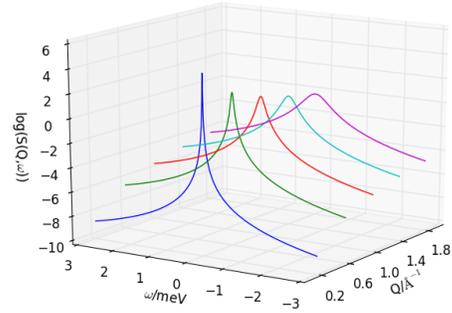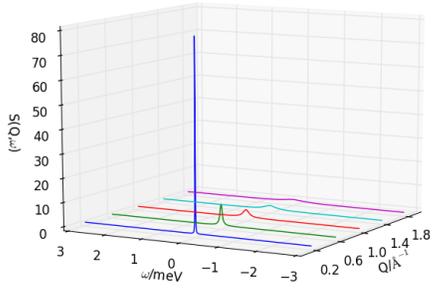
Dynamic structure factor, S(Q, ω) for a Brownian diffusion motion with a self-diffusion coefficient D = 0.1 meV Å² ≡ 1.5192 × 10⁻⁹ m2/s. (Right) Log-representation.

Limits

This model can be used to represent the translational component of the dynamic structure factor at low Q, where the corresponding investigated distance in real space are large enough to imply a large number of jumps. Deviations are expected at high Q values, where specific details of the jump mechanism start to be observable. In this case, such details need to be introduced in the scattering model, as e.g. in the Chudley-Elliott or Singwi-Sjolander models.

References

M. Bée: "Quasielastic Neutron Scattering: Principles and Applications in Solid State Chemistry, Biology and Materials Science", Adam-Hilger, Bristol, 1988; chapter 5.1.

R. Hempelman: "Quasielastic Neutron Scattering and Solid State Diffusion", Clarendon Press, Oxford, 2000; chapter 5.2

4) Long range translational diffusion (jump-diffusion):

   Y = QENS_JumpDiffusionTranslation (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2/3)} \cdot \frac{1}{\pi} \cdot \frac{\Gamma}{(\omega - P_2)^2 + \Gamma^2}$$

$$\Gamma = \frac{DQ^2}{1 + \tau DQ^2}$$

Jump-diffusion model with *P5* = [D] (self-diffusion constant in Å²·meV) and *P6* = [τ] (residence time in meV⁻¹). Fixing *P6* = 0, this model reduces to the random walk model.

5) Chudley-Elliot model for long range translational diffusion :

   Y = QENS_ChudleyElliot (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2 / 3)} \cdot \frac{1}{\pi} \cdot \frac{\Gamma}{(\omega - P_2)^2 + \Gamma^2}$$

$$\Gamma = \frac{1}{\tau}\left[1 - \frac{\sin(Ql)}{Ql}\right]$$

Jump-diffusion model with $P5$ = [l] (jump distance in Å) and $P6$ = [τ] (residence time in meV^{-1}).

6) Jump model among N equivalent sites in a circle:

   Y = QENS_JumpsNsites (X, P1, P2, P3, P4, P5, P6, P7)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2 / 3)} \cdot \left[A_0(Q) \cdot \delta(\omega) + \sum_{i=1}^{N-1} A_i(Q) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2}\right]$$

$$A_i(Q) = \frac{1}{N}\sum_{j=1}^{N} j_0(Qr_j)\cos\left(\frac{2ij\pi}{N}\right)$$

$$r_j = 2R\sin\left(\frac{j\pi}{N}\right)$$

$$\Gamma_i = \frac{1}{\tau_i} = \frac{2}{\tau}\sin^2\left(\frac{i\pi}{N}\right)$$

Parameters:

   $P5$: Number of sites [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

   $P6$: Radius of the circle [R] (Å).

   $P7$: [τ^{-1}] (meV).

7) Jump model among N equivalent sites in a circle with a log-Gaussian distribution of relaxation times:

   Y = QENS_JumpsNsitesLogNormDist (X, P1, P2, P3, P4, P5, P6, P7, P8)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2 / 3)} \cdot \left[A_0(Q) \cdot \delta(\omega) + \sum_{i=1}^{N-1} A_i(Q) \cdot \left(\sum_{j=1}^{L} g_j \cdot \frac{1}{\pi} \frac{\Gamma_{i,j}}{(\omega - P_2)^2 + \Gamma_{i,j}^2}\right)\right]$$

$$A_i(Q) = \frac{1}{N}\sum_{j=1}^{N} j_0(Qr_j)\cos\left(\frac{2ij\pi}{N}\right)$$

$$r_j = 2R \sin\left(\frac{j\pi}{N}\right)$$

$$\Gamma_i = \frac{1}{\tau_i} = \frac{2}{\tau} \sin^2\left(\frac{i\pi}{N}\right)$$

$$g_j = g(\ln \Gamma_{i,j}) = \frac{1}{\sigma\sqrt{2\pi}} \cdot \exp\left[-\frac{1}{2\sigma^2} \ln^2\left(\frac{\Gamma_{i,j}}{\Gamma_i}\right)\right]$$

Based on the rotation distribution model (A. Chahid, A. Alegría, J. Colmenero; *Macromolecules* **27**, 3282-3288 (1994)) that considers a distribution of jumping rates. For each jumping distance, instead of a single Γ_i value, a distribution of HWHMs is used. The distribution will be represented by L values of the HWHM ($\Gamma_{l,j}$) with associated weights g_j taken from a log-Gaussian distribution of standard deviation σ and normalized such that $\sum_{j=1}^{L} g_j = 1$. The $\Gamma_{l,j}$ are chosen equally spaced in logarithmic scale in the range $\left[\exp(-\sigma\sqrt{-2\ln A_{min}}), \exp(\sigma\sqrt{-2\ln A_{min}})\right]$, where A_{min} is the cut-off chosen for the value of the distribution function with respect to its maximum. At present the code uses L =21 and A_{min}=0.1.

Parameters:

P5: Number of sites [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

P6: Radius of the circle [R] (Å).

P7: [τ^{-1}] (meV).

P8: [σ] (adim).

8) Isotropic rotational diffusion in a sphere of radius R:

    Y = QENS_IsotropicRotationalDiffusion (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot P_3 \cdot e^{(-P_4 Q^2/3)} \cdot \left[A_0(Q) \cdot \delta(\omega) + \sum_{i=1}^{\infty} A_i(Q) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2}\right]$$

$$A_i(Q) = (2i + 1)j_i^2(QR)$$

$$\Gamma_i = i(i + 1)D_R$$

In the code, the infinite sum is replaced by a sum over the first 10 terms.

Parameters:

P5: Radius of the sphere [R] (Å).

P6: Rotational diffusion constant [D_R] (meV).

9) Long-range translation + isotropic rotational diffusion in a sphere of radius R:

    Y = QENS_RotTrans (X, P1, P2, P3, P4, P5, P6, P7, P8)

$$Y = P_1 \cdot P_3 \cdot e^{\left(-\frac{P_4 Q^2}{3}\right)} \cdot \left(\frac{1}{\pi}\frac{\Gamma_T}{(\omega - P_2)^2 + \Gamma_T^2}\right) \otimes \left(A_0(Q) \cdot \delta(\omega) + \sum_{i=1}^{\infty} A_i(Q) \cdot \frac{1}{\pi}\frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2}\right)$$

$$\Gamma_T = \frac{D_T Q^2}{1 + \tau D_T Q^2}$$

$$A_i(Q) = (2i + 1)j_i^2(QR)$$

$$\Gamma_i = i(i + 1)D_R$$

Parameters:

P5: Self-diffusion constant [D] (Å²·meV)

P6: Residence time [τ] (meV^{-1})

P7: Radius of the sphere [R] (Å).

P8: Rotational diffusion constant [D$_R$] (meV).

10) Gaussian model for localized translational motion (3D):

Y = QENS_GaussianModel3D (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot P_3 \cdot e^{\left(-\frac{P_4 Q^2}{3}\right)} \cdot \left(A_0(Q) \cdot \delta(\omega) + \sum_{i=1}^{\infty} A_i(Q) \cdot \frac{1}{\pi}\frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2}\right)$$

$$\Gamma_i = \frac{i D_T}{\langle u_x^2 \rangle}$$

$$A_i(Q) = \exp(-Q^2 \langle u_x^2 \rangle)\frac{(Q^2 \langle u_x^2 \rangle)^i}{i!}$$

Gaussian model for localized translational motion in 3D proposed in F. Volino, J.-C. Perrin, and S. Lyonnard; *J. Phys. Chem. B* **110**, 11217-11223 (2006). As said in the abstract of the paper, it can advantageously replace the previous model of diffusion inside a sphere with an impermeable surface (F. Volino and A. J. Dianoux, *Mol. Phys.* **41**, 271-279 (1980), as it is simpler. It may also be more appropriate when the confinement is defined by soft, ill-defined boundaries. Implemented using eqns. (37) and (27a-27b) in Volino's paper. Supposing a particle that can move along the direction *x* about a fixed point taken as the origin and being u_x the displacement from the origin, the model assumes that u_x is a Gaussian random variable with variance $\langle u_x^2 \rangle$. For the 3D case, the model assumes also $\langle u_x^2 \rangle = \langle u_y^2 \rangle = \langle u_z^2 \rangle$.

In the present code, the number of terms in the infinite sum is limited to 100. Volino et al. indicate that, as a rule of thumb, the number of terms n_{max} to be considered in practical calculations must be (much) larger than $Q^2 \langle u_x^2 \rangle$. Therefore any user employing this model should check that this condition is respected.

Note that the code uses a relative large number of lorentzians and double precision in order to deal with the large values that the argument $(Q^2\langle u_x^2\rangle)^i$ can take, so the fitting (specially 2D fits) can take a considerable time.

Parameters:

P5: Self-diffusion constant [D] (Å²·meV)

P6: Variance [$\langle u_x^2\rangle$] (Å²)

Combined functions

In principle, all the functions above can be added to produce more complex functions. Normally this will require linking some parameters between them. For example, if we have a system with two types of atoms (one not moving and another executing a simple motion that can be represented by a single Lorentzian), we could combine the functions QENS_delta and QENS_lorentz as shown in the figure below.
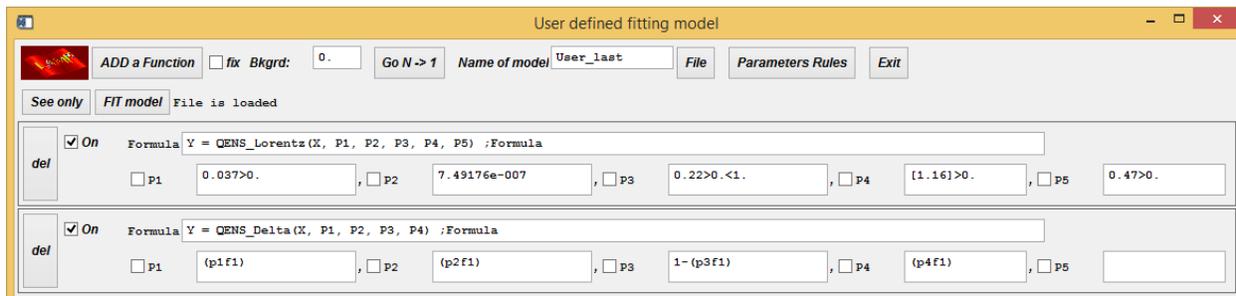
As shown by the example, we need to use the same scaling parameter for both functions (so the scaling factor for the delta function = (P1F1), i.e. the parameter P1 used in the first function, QENS_Lorentz). The same happens with the position and the Debye-Waller terms, so we also have P2 (delta) = (P2F1) and P4 (delta) = (P4F1). Finally, as we have two population of atoms, we need to specify the limits for P3 (lorentzian) between 0 and 1 and set P3 (delta) = 1-(P3F1).

Note that when linking two parameters, the parenthesis are compulsory, so writing P1 (delta) = P1F1 will fail!

Problems

While this approach works and allows to create complex combinations using the library of models available, we have found that when using more than one function the fit is less robust and often converges to bad solutions. This is particularly evident when using the delta function (possibly because the derivatives used by the fitting motor are discontinuous). Avoiding using the delta as the reference (first) function and tying the parameters of the delta to those of other function (and not the reverse) can help with this, but probably the fit will still require extensive manual intervention.

A practical solution to this problem is to write a new function that combines two or more of the simple models, so the fitting motor will call a single function and there is no need to tie any parameters together. There are already several combined functions that have been added to the QENS library, as shown below. The main difference with the previous models is that in the combined functions the parameter P3 is not related any more to the 'weight' of the function to the total sum. Thus they are normally intended to be used isolated and not in combination with other functions, but it is up to the user to decide if he needs to add a second function and how the parameters must be interpreted or tied. For example, in some cases it could be useful to combine a QENS_DeltaLorentz to describe the quasielastic spectrum and an additional QENS_Lorentz to describe some independent inelastic feature at $\omega \neq 0$.

The available functions are described below. Additional functions can be added upon request.

101) Delta + Lorenzian: Y = QENS_DeltaLorentz (X, P1, P2, P3, P4, P5)

$$Y = P_1 \cdot e^{(-P_4 Q^2/3)} \cdot \left[P_3 \cdot \delta(\omega) + (1 - P_3) \cdot \frac{1}{\pi} \cdot \frac{P_5}{(\omega - P_2)^2 + P_5^2} \right]$$

102) Delta + 2 Lorenzians: Y = QENS_DeltaLorentz2 (X, P1, P2, P3, P4, P5, P6, P7)

$$Y = P_1 \cdot e^{(-P_4 Q^2/3)} \cdot \left[P_3 \cdot \delta(\omega) + P_5 \cdot \frac{1}{\pi} \cdot \frac{P_6}{(\omega - P_2)^2 + P_6^2} + (1 - P_3 - P_5) \cdot \frac{1}{\pi} \cdot \frac{P_7}{(\omega - P_2)^2 + P_7^2} \right]$$

Caution: The sum of the intensities of the delta and the two Lorenzians should be 1. However, although the user can limit the parameters to be in the range [0, 1], the function does not check that P3+P5 < 1. In this case, the second Lorentzian will be subtracted (as 1−P3-P5 < 0).

102b) Delta + 2 Lorenzians: Y = QENS_DeltaLorentz2b (X, P1, P2, P3, P4, P5, P6, P7)

$$Y = P_1 \cdot e^{(-P_4 Q^2/3)} \cdot \left[(1 - P_3 - P_5) \cdot \delta(\omega) + P_3 \cdot \frac{1}{\pi} \cdot \frac{P_6}{(\omega - P_2)^2 + P_6^2} + P_5 \cdot \frac{1}{\pi} \cdot \frac{P_7}{(\omega - P_2)^2 + P_7^2} \right]$$

As the previous one, but with the two intensity parameters defining both Lorentzians, instead of the elastic peak and one lorentzian

103) Delta + ChudleyElliot: Y = QENS_DeltaChudleyElliot (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot e^{(-P_4 Q^2/3)} \cdot \left[P_3 \cdot \delta(\omega) + (1 - P_3) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_{CE}}{(\omega - P_2)^2 + \Gamma_{CE}^2} \right]$$

Parameters:

  $P1$: Intensity scaling (arbitrary units).

  $P2$: Center of the functions (meV).

  $P3$: Fraction of immobile protons [f_0].

  $P4$: [$\langle u^2 \rangle$] (Å²).

  $P5$: Characteristic jump distance [l_{CE}] (Å).

  $P6$: Mean residence time [τ_{CE}] (meV^{-1}) of restricted water

104) Water in biomolecular systems (Model described in F. Natali, Y. Gerelli, C. Stelletta and J. Peters, *AIP Conf. Proc.* **1518**, 551 (2013))**:**

  Y = QENS_Natali (X, P1, P2, P3, P4, P5, P6, P7, P8, P9, P10, P11, P12, P13, P14, P15)

$$Y = P_1 \cdot e^{\left(-\frac{P_4 Q^2}{3}\right)} \cdot \begin{bmatrix} P_3 \cdot \delta(\omega) + \\ P_5 \cdot S_1^{\text{RotTrans}}(Q, \omega, D_{w1}, \tau_{w1}, R_{w1}, D_{R,w1}) + \\ P_{10} \cdot S_2^{\text{RotTrans}}(Q, \omega, D_{w2}, \tau_{w2}, R_{w2}, D_{R,w2}) + \\ (1 - P_3 - P_5 - P_{10}) \cdot \frac{1}{\pi} \cdot \frac{P_{15}}{(\omega - P_2)^2 + P_{15}^2} \end{bmatrix}$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: Fraction of immobile protons [f_0].

P4: [$\langle u^2 \rangle$] (Å^2).

P5: Fraction of protons contributing to first roto-translational lorentzian [f_1].

P6: Self-diffusion constant [D] ($\text{Å}^2 \cdot \text{meV}$) of free water.

P7: Residence time [τ] (meV^{-1}) of free water.

P8: Radius of the sphere [R] (Å) of rotation for free water (fixed to 1.0 Å for water).

P9: Rotational diffusion constant [D_R] (meV) of free water.

P10: Fraction of protons contributing to second roto-translational lorentzian [f_2].

P11: Self-diffusion constant [D] ($\text{Å}^2 \cdot \text{meV}$) of restricted water.

P12: Residence time [τ] (meV^{-1}) of restricted water.

P13: Radius of the sphere [R] (Å) of rotation for restricted water (fixed to 1.0 Å for water).

P14: Rotational diffusion constant [D_R] (meV) of restricted water.

P15: HWHM (meV) of 3rd Lorentzian related to a faster relaxation.

Caution: As for QENS_DeltaLorentz2, nothing ensures that P3+P5+P10 < 1.

105) Water in biomolecular systems (Simplified model: No water rotations)**:**

   Y = QENS_Iri (X, P1, P2, P3, P4, P5, P6, P7, P8, P9, P10, P11)

$$Y = P_1 \cdot e^{\left(-\frac{P_4 Q^2}{3}\right)} \cdot \begin{bmatrix} P_3 \cdot \delta(\omega) + \\ P_5 \cdot S_1^{\text{Trans}}(Q, \omega, D_{w1}, \tau_{w1}) + \\ P_8 \cdot S_2^{\text{Trans}}(Q, \omega, D_{w2}, \tau_{w2}) + \\ (1 - P_3 - P_5 - P_8) \cdot \frac{1}{\pi} \cdot \frac{P_{11}}{(\omega - P_2)^2 + P_{11}^2} \end{bmatrix}$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: Fraction of immobile protons [f_0].

P4: [$\langle u^2 \rangle$] (Å^2).

P5: Fraction of protons contributing to first roto-translational lorentzian [f_1].

P6: Self-diffusion constant [D] ($\text{Å}^2 \cdot \text{meV}$) of free water.

P7: Residence time [τ] (meV^{-1}) of free water.

P8: Fraction of protons contributing to second roto-translational lorentzian [f$_2$].

P9: Self-diffusion constant [D] (Å^2·meV) of restricted water.

P10: Residence time [τ] (meV^{-1}) of restricted water.

P11: HWHM (meV) of 3rd Lorentzian related to a faster relaxation.

Caution: Again, nothing ensures that P3+P5+P8 < 1.

106) Water in biomolecular systems (Simplified model: Only 1 roto-translational component)**:**

Y = QENS_Iri2 (X, P1, P2, P3, P4, P5, P6, P7, P8, P9, P10)

$$Y = P_1 \cdot e^{\left(-\frac{P_4 Q^2}{3}\right)} \cdot \left[\begin{array}{c} P_3 \cdot \delta(\omega) + \\ P_5 \cdot S_1^{\text{RotTrans}}\left(Q, \omega, D_{w1}, \tau_{w1}, R_{w1}, D_{R,w1}\right) + \\ (1 - P_3 - P_5) \cdot \frac{1}{\pi} \cdot \frac{P_{10}}{(\omega - P_2)^2 + P_{10}^2} \end{array} \right]$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: Fraction of immobile protons [f$_0$].

P4: [⟨u^2⟩] (Å^2).

P5: Fraction of protons contributing to first roto-translational lorentzian [f$_1$].

P6: Self-diffusion constant [D] (Å^2·meV) of free water.

P7: Residence time [τ] (meV^{-1}) of free water.

P8: Radius of the sphere [R] (Å) of rotation for free water (fixed to 1.0 Å for water).

P9: Rotational diffusion constant [D$_R$] (meV) of free water.

P10: HWHM (meV) of 3rd Lorentzian related to a faster relaxation.

Caution: Again, nothing ensures that P3+P5 < 1.

107) Ionic liquids (Convolution of one lorentzian representing long-range translation + one localized component)**:**

Y = QENS_IL1 (X, P1, P2, P3, P4, P5, P6)

$$Y = P_1 \cdot e^{\left(-\frac{P_3 Q^2}{3}\right)} \cdot \left[P_4 \cdot \frac{1}{\pi} \cdot \frac{P_5}{(\omega - P_2)^2 + P_5^2} + (1 - P_4) \cdot \frac{1}{\pi} \cdot \frac{(P_5 + P_6)}{(\omega - P_2)^2 + (P_5 + P_6)^2} \right]$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å^2).

P4: EISF of localized component (range [0, 1]).

P5: HWHM (meV) of translational component

P6: HWHM (meV) of localized component.

108) Ionic liquids (Convolution of one lorentzian representing long-range translation + two localized components)**:**

   Y = QENS_IL2 (X, P1, P2, P3, P4, P5, P6, P7, P8)

$$Y = P_1 \cdot e^{\left(-\frac{P_3 Q^2}{3}\right)} \cdot \left[\begin{array}{c} P_4 \cdot P_5 \cdot \dfrac{1}{\pi} \cdot \dfrac{P_6}{(\omega - P_2)^2 + P_6^2} + \\[2mm] (1 - P_4) \cdot P_5 \cdot \dfrac{1}{\pi} \cdot \dfrac{(P_6 + P_7)}{(\omega - P_2)^2 + (P_6 + P_7)^2} + \\[2mm] P_4 \cdot (1 - P_5) \cdot \dfrac{1}{\pi} \cdot \dfrac{(P_6 + P_8)}{(\omega - P_2)^2 + (P_6 + P_8)^2} + \\[2mm] (1 - P_4) \cdot (1 - P_5) \cdot \dfrac{1}{\pi} \cdot \dfrac{(P_6 + P_7 + P_8)}{(\omega - P_2)^2 + (P_6 + P_7 + P_8)^2} \end{array} \right]$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å^2).

P4: EISF of 1st localized component (range [0, 1]).

P5: EISF of 2nd localized component (range [0, 1]).

P6: HWHM (meV) of translational component

P7: HWHM (meV) of 1st localized component.

P8: HWHM (meV) of 2nd localized component

109) Ionic liquids (Convolution of a jump-diffusion translation with a confined translation (Volino's model) and a general localized motion)**:**

   Y = QENS_IL3 (X, P1, P2, P3, P4, P5, P6, P7, P8, P9)

The model can be written as:

$$S(Q, \omega) \approx \mathcal{L}(\Gamma_T) \otimes \left[A_0^V(Q)\delta(\omega) + \sum_{i=1}^{\infty} A_i^V(Q)\mathcal{L}(\Gamma_i^V) \right] \otimes \left[A_0^L(Q) + \left(1 - A_0^L(Q)\right)\mathcal{L}(\Gamma_L(Q)) \right],$$

with

$$\Gamma_T = \frac{DQ^2}{1+\tau DQ^2}$$ (see long range translational diffusion (jump-diffusion) model),

$$A_i^V(Q) = \exp(-Q^2 \langle u_x^2 \rangle) \frac{(Q^2 \langle u_x^2 \rangle)^i}{i!}, \quad \Gamma_i^V = \frac{iD_T}{\langle u_x^2 \rangle}$$ (see Volino's Gaussian model for localized translational motion (3D)),

$A_0^L(Q)$ = Generic EISF for an arbitrary localized motion, and

$\Gamma_L(Q)$ = HWHM of the arbitrary localized motion.

Thus one has:

$$S(Q,\omega) \approx A_0^V(Q)A_0^L(Q)\mathcal{L}(\Gamma_T) +$$

$$A_0^L(Q)\sum_{i=1}^{\infty} A_i^V(Q)\mathcal{L}(\Gamma_i^V + \Gamma_T) +$$

$$A_0^V(Q)\left(1 - A_0^L(Q)\right)\mathcal{L}(\Gamma_L(Q) + \Gamma_T) +$$

$$\left(1 - A_0^L(Q)\right)\sum_{i=1}^{\infty} A_i^V(Q)\mathcal{L}(\Gamma_i^V + \Gamma_L(Q) + \Gamma_T)$$

And the coded function is:

$$Y = P_1 \cdot e^{\left(-\frac{P_3 Q^2}{3}\right)} \cdot \begin{bmatrix} A_0^V(Q) \cdot P_8 \cdot \dfrac{1}{\pi} \cdot \dfrac{\Gamma_T}{(\omega - P_2)^2 + \Gamma_T^2} + \\[2mm] P_8 \cdot \sum_{i=1}^{100} A_i^V(Q) \cdot \dfrac{1}{\pi} \cdot \dfrac{\left(\Gamma_i^V + \Gamma_T\right)}{(\omega - P_2)^2 + \left(\Gamma_i^V + \Gamma_T\right)^2} + \\[2mm] A_0^V(Q) \cdot (1 - P_8) \cdot \dfrac{1}{\pi} \cdot \dfrac{(P_9 + \Gamma_T)}{(\omega - P_2)^2 + (P_9 + \Gamma_T)^2} + \\[2mm] (1 - P_8) \cdot \sum_{i=1}^{100} A_i^V(Q) \cdot \dfrac{1}{\pi} \cdot \dfrac{\left(P_9 + \Gamma_i^V + \Gamma_T\right)}{(\omega - P_2)^2 + \left(P_9 + \Gamma_i^V + \Gamma_T\right)^2} \end{bmatrix}$$

with

$$\Gamma_T = \frac{P_4 Q^2}{1 + P_5 P_4 Q^2}$$

$$A_i^V(Q) = \exp(-Q^2 P_7) \frac{(Q^2 P_7)^i}{i!}$$

$$\Gamma_i^V = \frac{iP_6}{P_7}$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å²).

P4: [D] (self-diffusion constant in Å²·meV for the long-range translation).

P5: [τ] (residence time in meV⁻¹ for the long-range translation).

P6: [D_{conf}] (self-diffusion constant in Å²·meV for the confined translation).

P7: Variance [$\langle u_x^2 \rangle$] (Å²) in Volino's model for confined translation.

P8: EISF of localized component (range [0, 1]).

P9: HWHM (meV) of localized component.

110) Jump model among N equivalent sites in a circle + fraction of fixed atoms:

Y = QENS_Fixed_JumpsNsites (X, P1, P2, P3, P4, P5, P6, P7)

$$Y = P_1 \cdot e^{\left(-P_3 Q^2/3\right)} \cdot \left[[P_4 + (1 - P_4) \cdot A_0(Q)] \cdot \delta(\omega) + \sum_{i=1}^{N-1} (1 - P_4) \cdot A_i(Q) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2} \right]$$

$$A_i(Q) = \frac{1}{N} \sum_{j=1}^{N} j_0(Qr_j) \cos\left(\frac{2ij\pi}{N}\right)$$

$$r_j = 2R \sin\left(\frac{j\pi}{N}\right)$$

$$\Gamma_i = \frac{1}{\tau_i} = \frac{2}{\tau} \sin^2\left(\frac{i\pi}{N}\right)$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å²).

P4: Fraction of fixed atoms (range [0,1]).

P5: Number of sites [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

P6: Radius of the circle [R] (Å).

P7: [τ^{-1}] (meV).

111) 2 x Jump model among N equivalent sites in a circle + fraction of fixed atoms:

Y = QENS_Fixed_2JumpsNsites (X, P1, P2, P3, P4, P5, P6, P7, P8, P9, P10, P11)

$$Y = P_1 \cdot e^{(-P_3 Q^2/3)} \cdot \begin{bmatrix} \{(1 - P_4 - P_8) + P_4 \cdot A_0^I(Q) + P_8 \cdot A_0^{II}(Q)\} \cdot \delta(\omega) + \\ \sum_{i=1}^{N_1-1} P_4 \cdot A_i(Q) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_i}{(\omega - P_2)^2 + \Gamma_i^2} + \\ \sum_{j=1}^{N_2-1} P_8 \cdot A_j(Q) \cdot \frac{1}{\pi} \cdot \frac{\Gamma_j}{(\omega - P_2)^2 + \Gamma_j^2} \end{bmatrix}$$

$$A_i(Q) = \frac{1}{N_1} \sum_{k=1}^{N} j_0(Qr_k) \cos\left(\frac{2ik\pi}{N_1}\right) \quad ; \quad r_k = 2R_1 \sin\left(\frac{k\pi}{N_1}\right) \quad ; \quad \Gamma_i = \frac{1}{\tau_i} = \frac{2}{\tau_1} \sin^2\left(\frac{i\pi}{N_1}\right)$$

$$A_j(Q) = \frac{1}{N_2} \sum_{k=1}^{N} j_0(Qr_k) \cos\left(\frac{2ik\pi}{N_2}\right) \quad ; \quad r_k = 2R_2 \sin\left(\frac{k\pi}{N_2}\right) \quad ; \quad \Gamma_j = \frac{1}{\tau_j} = \frac{2}{\tau_2} \sin^2\left(\frac{j\pi}{N_2}\right)$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å²).

P4: Fraction of atoms contributing to first jump model (range [0,1]).

P5: Number of sites in first jump model [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

P6: Radius of the circle in first jump model[R] (Å).

P7: [τ^{-1}] for first jump model (meV).

P8: Fraction of atoms contributing to second jump model (range [0,1]).

P9: Number of sites in second jump model [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

P10: Radius of the circle in second jump model [R] (Å).

P11: [τ^{-1}] for second jump model (meV).

112) Jump model among N equivalent sites in a circle with a log-Gaussian distribution of relaxation times + fraction of fixed atoms:

Y = QENS_Fixed_JumpsNsitesLogNormDist (X, P1, P2, P3, P4, P5, P6, P7, P8)

$$Y = P_1 \cdot e^{(-P_3 Q^2/3)} \left[[P_4 + (1 - P_4) \cdot A_0(Q)]\delta(\omega) + \sum_{i=1}^{N-1} (1 - P_4) \cdot A_i(Q) \left(\sum_{j=1}^{L} g_j \cdot \frac{1}{\pi} \frac{\Gamma_{i,j}}{(\omega - P_2)^2 + \Gamma_{i,j}^2} \right) \right]$$

$$A_i(Q) = \frac{1}{N} \sum_{j=1}^{N} j_0(Qr_j) \cos\left(\frac{2ij\pi}{N}\right)$$

$$r_j = 2R \sin\left(\frac{j\pi}{N}\right)$$

$$\Gamma_i = \frac{1}{\tau_i} = \frac{2}{\tau} \sin^2\left(\frac{i\pi}{N}\right)$$

$$g_j = g\left(\ln \Gamma_{i,j}\right) = \frac{1}{\sigma\sqrt{2\pi}} \cdot \exp\left[-\frac{1}{2\sigma^2} \ln^2\left(\frac{\Gamma_{i,j}}{\Gamma_i}\right)\right]$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å^2).

P4: Fraction of fixed atoms (range [0,1]).

P5: Number of sites [N]. This parameter is converted to an integer by the function. Normally it is intended to be used only as a fixed parameter defining the number of sites and it should not be fitted.

P6: Radius of the circle [R] (Å).

P7: [τ^{-1}] (meV).

P8: [σ] (adim).

113) Delta + Translation + Confined translation (Volino's model) + Isotropic rotation:

  Y = QENS_Mix1 (X, P1, P2, P3, P4, P5, P6, P7, P8, P9, P10, P11, P12)

$$Y = P_1 \cdot e^{\left(-\frac{P_3 Q^2}{3}\right)} \cdot \begin{bmatrix} P_4 \cdot \delta(\omega) + \\ P_5 \cdot S_1^{\text{JumpTrans}}(Q, \omega, D_1, \tau_1) + \\ P_8 \cdot S_2^{\text{Volino}}(Q, \omega, D_2, u_2) + \\ (1 - P_4 - P_5 - P_8) \cdot S_3^{\text{IsoRot}}(Q, \omega, D_3, R_3) \end{bmatrix}$$

Parameters:

P1: Intensity scaling (arbitrary units).

P2: Center of the functions (meV).

P3: [$\langle u^2 \rangle$] (Å^2).

P4: Fraction of immobile protons [f_0].

P5: Fraction of protons contributing to first Lorentzian (long-range jump translational diffusion) [f_1].

P6 = [D] (self-diffusion constant in Å^2·meV)

P7 = [τ] (residence time in meV^{-1}).

P8: Fraction of protons contributing to second Lorentzian (confined translational diffusion, Volino) [f_2].

P9: Self-diffusion constant [D] (Å^2·meV).

P10: Variance [$\langle u_x^2 \rangle$] (Å^2).

P11: Radius of the sphere [R] (Å) for isotropic rotational diffusion.

P12: Rotational diffusion constant [D_R] (meV).

Caution: As for QENS_DeltaLorentz2, nothing ensures that P3+P5+P10 < 1.