The RIXS 2D map is defined as an intensity function of two variables, which are the energies of the incoming and outgoing photon, ω_i and ω_f respectively.

$$I_{\mu\nu}(\omega_i, \omega_f) = \sum_f |\langle \Psi_f | O_{\mu\nu}(\omega_i) | \Psi_i \rangle|^2 \delta((\omega_i - \omega_f) + E_i - E_f) \tag{1}$$

where $O_{\mu\nu}$ is the RIXS transition operator as function of ω_i

$$O_{\mu\nu}(\omega_i) = D^\dagger_\mu \frac{1}{\omega_i - \mathcal{H} + i\Gamma} D_\nu \tag{2}$$

in which $\mathcal{H}$ is the Hamiltonian with the core-hole potential. Now define the RIXS matrix element $\mathcal{M}_{f,\mu\nu}$ and expand it with the eigenstate $|\tilde{\Psi}_n\rangle$ of $\mathcal{H}$

$$\begin{aligned}
\mathcal{M}_{f,\mu\nu}(\omega_i) \equiv& \langle \Psi_f | O_{\mu\nu}(\omega_i) | \Psi_i \rangle \\
=& \langle \Psi_f | D^\dagger_\mu \frac{1}{\omega_i - \mathcal{H} + i\Gamma} D_\nu | \Psi_i \rangle \\
=& \sum_n \langle \Psi_f | D^\dagger_\mu \frac{1}{\omega_i - \mathcal{H} + i\Gamma} | \tilde{\Psi}_n \rangle \langle \tilde{\Psi}_n | D_\nu | \Psi_i \rangle \\
=& \sum_n \frac{\langle \Psi_f | D^\dagger_\mu | \tilde{\Psi}_n \rangle \langle \tilde{\Psi}_n | D_\nu | \Psi_i \rangle}{\omega_i - \tilde{\omega}_n + i\Gamma}
\end{aligned} \tag{3}$$

$\mathcal{M}_{f,\mu\nu}(\omega_i)$ is significant only when $\omega_i \approx \tilde{\omega}_n$ and $\tilde{\Psi}_n$ can be coupled to both Ψ_i and Ψ_f via the dipole operator D_μ. An example of is shown in Fig. 1. Note that each pole is asymmetric and has a fano lineshape due to interference effects. Then we can expect $I_{\mu\nu}(\omega_i, \omega_f)$ to have significant features only near

$$(\omega_i, \omega_f) = (\tilde{\omega}_n, \tilde{\omega}_n + (E_i - E_f)) \tag{4}$$

To obtain $I_{\mu\nu}(\omega_i, \omega_f)$ one needs to loop over the intermediate space n and the final-state space f.

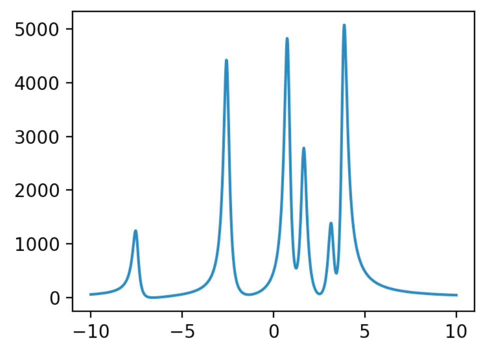

Figure 1: An example of the matrix element $\mathcal{M}_{f,\mu\nu}(\omega_i)$ as function of ω_i with 6 intermediate state n and a Γ of 0.2.

The matrix element $\langle\tilde{\Psi}_n|D_\nu|\Psi_i\rangle$ is simply the XAS matrix element. Then what is $\langle\Psi_f|D_\mu^\dagger|\tilde{\Psi}_n\rangle$?

$$\begin{aligned}
&\langle\tilde{\Psi}_n|D_\mu|\Psi_f\rangle \\
=&\langle 0|\prod_{j=1}^{N+1} a_{n_j}(\sum_c a_c^\dagger h\langle\psi_c|r_\mu|\psi_h\rangle)\prod_{k=1}^{N} a_{f_k}^\dagger h^\dagger|0\rangle \\
=&\sum_c\langle\psi_c|r_\mu|\psi_h\rangle(\det\Xi_{\{n_j\},\{f_k,c\}})^*
\end{aligned}\tag{5}$$

$\Xi_{\{n_j\},\{f_k,c\}}$ is a square sub-matrix that takes row $\{n_j\}$ and column $\{f_k,c\}$ from the transformation matrix Ξ. To understand how this sub-matrix is obtained, we may focus on the $f^{(1)}$ term, i.e., the single configurations, in both the initial- and intermediate-state picture. For the intermediate state, the $f^{(1)}$ terms of $|\tilde{\Psi}_n\rangle$ can be represented as

$$(n_1,n_2,\cdots,n_{N+1})=(1,2,\cdots,N,\tilde{c}_1)\tag{6}$$

In the final(initial)-state picture, the $f^{(1)}$ terms of $|\Psi_f\rangle$ can be represented as

$$(f_1,f_2,\cdots,f_N,c)=(1,2,\cdots,v_1-1,v_1+1,\cdots,N,c_1,c)\tag{7}$$

c can be any number and even in the gap between v_1-1 and v_1+1 as long as it doesn't coincide with other indices. If c is in the valence bands, that represent the emission from the occupied orbitals. These combination of rows and columns for the emission matrix element is shown in Fig. 2.

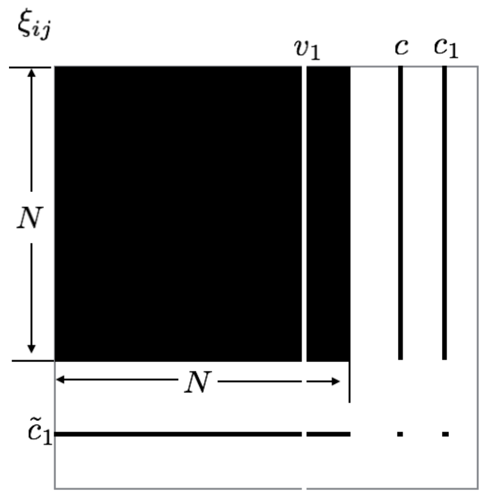

Figure 2: $E_c^{c_1 v_1, \tilde{c}_1}$

With these definitions, we may write down the matrix elements up to the single configurations ($f^{(1)}$) in a more specific form:

$$\langle \tilde{\Psi}_n | D_\mu | \Psi_f \rangle^{(1)} = \sum_c w_{c,\mu} (E_c^{c_1 v_1, \tilde{c}_1})^* \tag{8}$$

where

$$\begin{aligned} w_{c,\mu} &\equiv \langle \psi_c | r_\mu | \psi_h \rangle \\ E_c^{c_1 v_1, \tilde{c}_1} &\equiv \det \xi_{\{1,2,\cdots,N,\tilde{c}_1\},\{1,2,\cdots,v_1-1,v_1+1,\cdots,N,c_1,c\}} \end{aligned} \tag{9}$$

Then the RIXS matrix element at $f^{(1)}$ can be reduced to:

$$\mathcal{M}_{f,\mu\nu}(\omega_i) = \sum_{\tilde{c}_1} \frac{\sum_c E_c^{c_1 v_1, \tilde{c}_1} w_{c,\mu}^* \sum_{d \in \text{empty}} (A_d^{\tilde{c}_1})^* w_{d,\nu}}{\omega_i - \tilde{\omega}_{\tilde{c}_1} + i\Gamma} \tag{10}$$

Define a new sets of tensors as:

$$\begin{aligned} E_\mu^{c_1 v_1, \tilde{c}_1} &= \sum_c E_c^{c_1 v_1, \tilde{c}_1} w_{c,\mu}^* \\ A_\nu^{\tilde{c}_1} &= \sum_{d \in \text{empty}} A_d^{\tilde{c}_1} w_{d,\nu}^* \end{aligned} \tag{11}$$

The tensor $E_\mu^{c_1 v_1, \tilde{c}_1}$ can be obtained by altering two columns from $A_\nu^{\tilde{c}_1}$. To understand their relations, we introduce the exterior algebra to represent the tensors in terms of row vectors: $E_{\mu}^{c_1 v_1, \tilde{c}_1} = e_1 \wedge e_2 \wedge \cdots \wedge e_{N+1}$ and $A_{\nu}^{\tilde{c}_1} = a_1 \wedge a_2 \wedge \cdots \wedge a_{N+1}$. We may focus on the first row to understand the row indexing.

$$
\begin{aligned}
a_1 &= [\xi_{11}, \xi_{12}, \cdots, \xi_{1N}, \sum_{d \geq N} \xi_{1d} w^*_{d,\nu}] \\
e_1 &= [\xi_{11}, \xi_{12}, \cdots, \xi_{1,v_1-1}, \xi_{1,c_1}, \xi_{1,v_1+1}, \cdots, \xi_{1N}, \sum_{c \geq N, c \neq c_1} \xi_{1c} w^*_{c,\mu} + \xi_{1,v_1} w^*_{v_1,\mu}]
\end{aligned}
\tag{12}
$$

We can see that a_1 and e_1 differ at the v_1 (ξ_{1,v_1} v.s. ξ_{1,c_1}) and the last column, and therefore the tensor $E_{\mu}^{c_1 v_1, \tilde{c}_1}$ can be obtained by updating the columns of $A_{\nu}^{\tilde{c}_1}$. To this end, we now express $A_{\nu}^{\tilde{c}_1}$ in terms of the column vectors of the ξ matrix:

$$
A_{\nu}^{\tilde{c}_1} = \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_N \wedge \sum_{d > N} \Xi_d w^*_{d,\nu} \tag{13}
$$

where $\Xi_i \equiv \Xi_i^{\tilde{c}} \equiv (\xi_{1i}, \xi_{2i}, \cdots, \xi_{Ni}, \xi_{\tilde{c}i})^T$ and $\tilde{c}$ will be omitted hereafter. $E_{\mu}^{c_1 v_1, \tilde{c}_1}$ can be expressed as

$$
\begin{aligned}
E_{\mu}^{c_1 v_1, \tilde{c}_1} &= \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_{v_1-1} \wedge \Xi_{c_1} \wedge \Xi_{v_1+1} \wedge \cdots \wedge \Xi_N \\
&\quad \wedge \big[\sum_{c > N} \Xi_c w^*_{c,\mu} + (\Xi_{v_1} w^*_{v_1,\mu} - \Xi_{c_1} w^*_{c_1,\mu}) \big] \\
&= \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_{v_1-1} \wedge \Xi_{c_1} \wedge \Xi_{v_1+1} \wedge \cdots \wedge \sum_{c > N} \Xi_c w^*_{c,\mu} \\
&\quad + \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_{v_1-1} \wedge \Xi_{c_1} \wedge \Xi_{v_1+1} \wedge \cdots \wedge \Xi_{v_1} w^*_{v_1,\mu}
\end{aligned}
\tag{14}
$$

To facilitate the calculation, we slightly swap Ξ_{c_1} and Ξ_{v_1}.

$$
\begin{aligned}
E_{\mu}^{c_1 v_1, \tilde{c}_1} &= \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_{v_1-1} \wedge \Xi_{c_1} \wedge \Xi_{v_1+1} \wedge \cdots \wedge \Xi_N \wedge \sum_{c > N} \Xi_c w^*_{c,\mu} \\
&\quad - \big(\bigwedge_{j=1 \cdots N} \Xi_j \big) \wedge \Xi_{c_1} w^*_{v_1,\mu}
\end{aligned}
\tag{15}
$$

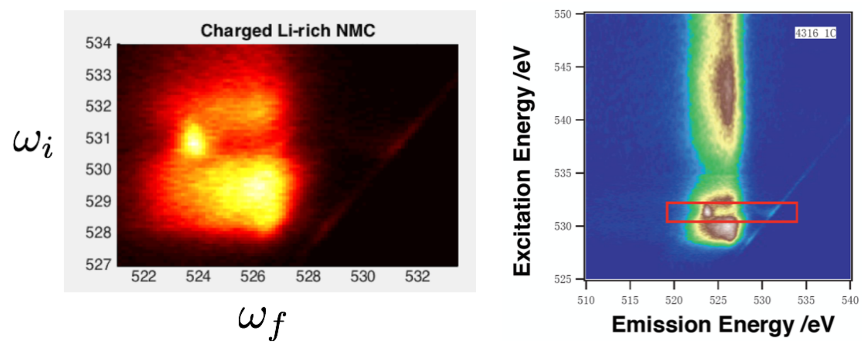

Figure 3: .

Now define two reference determinants for the updating calculations:

$$
\begin{aligned}
P &\equiv \Xi_1 \wedge \Xi_2 \wedge \cdots \wedge \Xi_{v_1-1} \wedge \Xi_{v_1} \wedge \Xi_{v_1+1} \wedge \cdots \wedge \Xi_N \wedge \sum_{c>N} \Xi_c w^*_{c,\mu} \\
&= (\bigwedge_{j=1\cdots N} \Xi_j) \wedge \sum_{c>N} \Xi_c w^*_{c,\mu} \qquad (16) \\
Q &\equiv (\bigwedge_{j=1\cdots N+1} \Xi_j)
\end{aligned}
$$

The first term in Eq. (15) can be obtained from P by replacing Ξ_{v_1} with Ξ_{c_1} while the second term can be obtained by from Q by replacing Ξ_{N+1} with Ξ_{c_1}. Then these determinants can be found by the previous method using the so-called ξ-matrix. A slight difference here is that we are now dealing with column vectors but not row vectors. Assume

$$P_c = (P_1, P_2, \cdots, P_{N+1})(\zeta_{c1}, \zeta_{c2}, \cdots, \zeta_{c,N+1})^T \tag{17}$$

then the ξ-matrix can be obtained by.

$$(\zeta_{c1}, \zeta_{c2}, \cdots, \zeta_{c,N+1})^T = P^{-1} P_c \tag{18}$$