Resonant inelastic x-ray scattering studies of elementary excitations

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Appendix A

QED

This appendix contains some details of the derivation of the higher-order electron-photon coupling Hamiltonian.

A.1 H_Ψ to second order

Below the components of $H_\Psi = \Omega H_\alpha \Omega^{-1} + i\hbar(\partial_t \Omega)\Omega^{-1}$ are calculated. First,

$$\Omega H_\alpha \Omega^{-1} = H_\alpha + \frac{1}{2}\left[\tilde{D}^2, H_\alpha\right] = H_\alpha + \frac{1}{2}\left[\tilde{D}^2, -e\phi\right]. \tag{A.1}$$

where in the last step only terms up to $\mathcal{O}(m^{-2})$ are kept, and where

$$\begin{aligned}(2mc)^2\left[\tilde{D}^2, \phi\right] &= \sigma^i\sigma^j\left(D_iD_j\phi - \phi D_iD_j\right) = \sigma^i\sigma^j\left(D_i(\partial_j\phi) + D_i\phi D_j - \phi D_iD_j\right)\\ &= \sigma^i\sigma^j\left(\partial_i(\partial_j\phi) + (\partial_j\phi)D_i + (\partial_i\phi)D_j\right) = \partial_i^2\phi + 2(\partial_i\phi)D_i\\ &= -\rho/\epsilon_0 + \frac{2i}{\hbar}(\nabla\phi)\cdot(\mathbf{p}+e\mathbf{A}).\end{aligned} \tag{A.2}$$

Second,

$$\begin{aligned}i\hbar\partial_t\Omega &= \frac{i\hbar}{2(2mc)^2}\partial_t\left[(\mathbf{p}+e\mathbf{A})^2 + e\hbar\boldsymbol{\sigma}\cdot\mathbf{B}\right]\\ &= \frac{ie\hbar}{2(2mc)^2}\left[(\partial_t\mathbf{A})\cdot(\mathbf{p}+e\mathbf{A}) + (\mathbf{p}+e\mathbf{A})\cdot(\partial_t\mathbf{A}) + \hbar\boldsymbol{\sigma}\cdot(\partial_t\mathbf{B})\right]\\ &= \frac{ie\hbar}{2(2mc)^2}\left[2(\partial_t\mathbf{A})\cdot(\mathbf{p}+e\mathbf{A}) + \hbar\boldsymbol{\sigma}\cdot(\partial_t\mathbf{B})\right]\end{aligned} \tag{A.3}$$

Putting things together, one finds

$$\begin{aligned} H_\Psi &= H_\alpha - \frac{1}{2}\frac{e\hbar^2\rho}{(2mc)^2\epsilon_0} - \frac{ie\hbar}{(2mc)^2}\mathbf{E}\cdot(\mathbf{p}+e\mathbf{A}) + \frac{ie\hbar^2}{2(2mc)^2}\boldsymbol{\sigma}\cdot(\partial_t\mathbf{B}) \\ &= \frac{(\mathbf{p}+e\mathbf{A})^2}{2m} + \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B} - e\phi + \frac{e\hbar}{(2mc)^2}\boldsymbol{\sigma}\cdot\mathbf{E}\times(\mathbf{p}+e\mathbf{A}) + \frac{1}{2}\frac{e\hbar^2\rho}{(2mc)^2\epsilon_0} \\ &\quad - \frac{ie\hbar}{2(2mc)^2}\hbar\boldsymbol{\sigma}\cdot(\partial_t\mathbf{B}). \end{aligned} \tag{A.4}$$

Note that the $\mathbf{E}\times\mathbf{p}$ term is not Hermitian for dynamic fields, because $\mathbf{E}$ and $\mathbf{p}$ do not commute in that case. The 'imaginary term' in H_Ψ solves this issue: it can be rewritten as

$$\begin{aligned} -\frac{ie\hbar}{2(2mc)^2}\hbar\sigma^k(\partial_t B^k) &= \frac{ie\hbar^2}{2(2mc)^2}\epsilon^{ijk}\sigma^k(\partial_i E^j) = \frac{ie\hbar^2}{2(2mc)^2}\epsilon^{ijk}\sigma^k(\partial_i E^j - E^j\partial_i) \\ &= -\frac{e\hbar}{2(2mc)^2}\epsilon^{ijk}\sigma^k(-i\hbar\partial_i E^j + E^i(-i\hbar)\partial_j) \\ &= -\frac{e\hbar}{2(2mc)^2}\boldsymbol{\sigma}\cdot(\mathbf{p}\times\mathbf{E}+\mathbf{E}\times\mathbf{p}) \end{aligned} \tag{A.5}$$

With this substitution, H_Ψ assumes the form of Eq. (2.22).

A.2 H_Ψ to third order

This section contains the derivation of H_Ψ to order $\mathcal{O}(m^{-3})$.

Starting point is Eq. (2.7):

$$\beta(x) = \tilde{D}\alpha(x) + \tilde{D}_0\tilde{D}\alpha(x) + \tilde{D}_0^2\tilde{D}\alpha(x). \tag{A.6}$$

Substitution in Eq. (2.5) gives the equation for $\alpha(x)$:

$$\tilde{D}_0\alpha(x) + \tilde{D}\left(1+\tilde{D}_0+\tilde{D}_0^2\right)\tilde{D}\alpha(x) = 0. \tag{A.7}$$

Every $\tilde{D}_0$ is commuted to the right:

$$\begin{aligned} 0 &= \left[\tilde{D}_0 + \tilde{D}^2 + \tilde{D}\left(1+\tilde{D}_0\right)\left(\tilde{D}\tilde{D}_0 + \frac{ie\hbar}{(2mc)^2c}\sigma^i E^i\right)\right]\alpha(x) \\ &= \Bigg[\tilde{D}_0 + \tilde{D}^2 + \tilde{D}\left(\tilde{D}\tilde{D}_0 + \frac{ie\hbar}{(2mc)^2c}\sigma^i E^i\right) \\ &\quad + \tilde{D}\left(\left(\tilde{D}\tilde{D}_0 + \frac{ie\hbar}{(2mc)^2c}\sigma^i E^i\right)\tilde{D}_0 + \tilde{D}_0\frac{ie\hbar}{(2mc)^2c}\sigma^i E^i\right)\Bigg]\alpha(x) \end{aligned} \tag{A.8}$$

Using

$$\left[\tilde{D}_0, E^i\right] = \frac{-i\hbar}{2mc}(\partial_0 E^i), \tag{A.9}$$

one obtains

$$-\tilde{D}_0\alpha(x) = \left[\tilde{D}^2\left(1+\tilde{D}_0+\tilde{D}_0^2\right)+\tilde{D}\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i\left(1+2\tilde{D}_0\right)\right.$$
$$\left.+\tilde{D}\frac{e\hbar^2\sigma^i(\partial_0E^i)}{(2mc)^3c}\right]\alpha(x). \tag{A.10}$$

$\tilde{D}_0\alpha(x)$ is replaced by the second order Schrödinger equation, and only terms that contribute to the Schrödinger equation at order $\mathcal{O}(m^{-3})$ are considered:

$$-\tilde{D}_0\alpha(x) = \left[\tilde{D}^2\left(1-\tilde{D}^2\right)+\tilde{D}\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i+\tilde{D}\frac{e\hbar^2\sigma^i(\partial_0E^i)}{(2mc)^3c}\right]\alpha(x). \tag{A.11}$$

The normalization condition becomes

$$\int d^3x\,\left[\alpha(x)^\dagger\alpha(x)+\left\{\left(1+\tilde{D}_0+\tilde{D}_0^2\right)\tilde{D}\alpha(x)\right\}^\dagger\left\{\left(1+\tilde{D}_0+\tilde{D}_0^2\right)\tilde{D}\alpha(x)\right\}\right]=1. \tag{A.12}$$

Up to order $\mathcal{O}(m^{-3})$, this is

$$\begin{aligned}
1 &= \int d^3x\,\left[\alpha(x)^\dagger+\alpha(x)^\dagger\tilde{D}^2+\left\{\tilde{D}_0\tilde{D}\alpha(x)\right\}^\dagger\tilde{D}+\alpha(x)^\dagger\tilde{D}\tilde{D}_0\tilde{D}\right]\alpha(x)\\
&= \int d^3x\,\left[\alpha(x)^\dagger+\alpha(x)^\dagger\tilde{D}^2+\left\{\left(\tilde{D}\tilde{D}_0+\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i\right)\alpha(x)\right\}^\dagger\tilde{D}\right.\\
&\quad\left.+\alpha(x)^\dagger\tilde{D}\left(\tilde{D}\tilde{D}_0+\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i\right)\right]\alpha(x)\\
&= \int d^3x\;\alpha(x)^\dagger\left[1+\tilde{D}^2+\left(\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i\right)^\dagger\tilde{D}+\tilde{D}\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i\right]\alpha(x)\\
&= \int d^3x\;\alpha(x)^\dagger\left[1+\tilde{D}^2+\frac{e\hbar}{(2mc)^2c}\left[\tilde{D},i\sigma^iE^i\right]\right]\alpha(x),
\end{aligned} \tag{A.13}$$

where the commutator

$$\left[\tilde{D},i\sigma^iE^i\right]=\frac{\hbar}{2mc}\left(\sigma^i\sigma^j(\partial_iE^j)-2i\epsilon^{ijk}\sigma^kE^iD_j\right) \tag{A.14}$$

is hermitian. The renormalization operator becomes

$$\Omega(x)=1+\frac{1}{2}\left(\tilde{D}^2+\frac{e\hbar}{(2mc)^2c}\left[\tilde{D},i\sigma^iE^i\right]\right)=\Omega(x)^\dagger, \tag{A.15}$$

so that to order $\mathcal{O}(m^{-3})$

$$\Omega(x)^{-1}=1-\frac{1}{2}\left(\tilde{D}^2+\frac{e\hbar}{(2mc)^2c}\left[\tilde{D},i\sigma^iE^i\right]\right). \tag{A.16}$$

With this renormalization operator, one can obtain the Schrödinger equation for the normalized wave function $\Psi(x)$. Its Hamiltonian H_Ψ consists of the parts

$$i\hbar(\partial_t\Omega)\Omega^{-1} = \frac{ie\hbar}{2(2mc)^2}\left\{2(\partial_t\mathbf{A})\cdot(\mathbf{p}+e\mathbf{A})+\hbar\boldsymbol{\sigma}\cdot(\partial_t\mathbf{B})\right\} + \frac{e}{2c}\left(\frac{-i\hbar}{2mc}\right)^3\left(\sigma^i\sigma^j(\partial_t\partial_i E^j) - 2i\epsilon^{ijk}\sigma^k(\partial_t E^i D_j)\right) \tag{A.17}$$

and

$$\begin{aligned}\Omega H_\alpha\Omega^{-1} &= 2mc^2\left(-\frac{e\phi}{2mc^2}+\tilde{D}^2-\tilde{D}^4+\tilde{D}\frac{ie\hbar}{(2mc)^2c}\sigma^iE^i+\tilde{D}\frac{e\hbar^2\sigma^i(\partial_0E^i)}{(2mc)^3c}\right)\\ &+\left[\frac{1}{2}\left(\tilde{D}^2+\frac{e\hbar}{(2mc)^2c}\left[\tilde{D},i\sigma^iE^i\right]\right),-e\phi+2mc^2\tilde{D}^2\right]\\ &= -e\phi+\frac{(\mathbf{p}+e\mathbf{A})^2}{2m}+\frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}-\frac{1}{2mc^2}\left(\frac{(\mathbf{p}+e\mathbf{A})^2}{2m}+\frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)^2\\ &-e\left(\frac{-i\hbar}{2mc}\right)^2\sigma^j\sigma^iD_jE^i-e\left(\frac{-i\hbar}{2mc}\right)^3\sigma^j\sigma^iD_j(\partial_0E^i)\\ &-\left(\frac{-i\hbar}{2mc}\right)^2\frac{e}{2}\left(\partial_i^2\phi+2(\partial_i\phi)D_i\right)-\frac{e^2\hbar}{2(2mc)^2c}\left[\left[\tilde{D},i\sigma^iE^i\right],\phi\right],\end{aligned} \tag{A.18}$$

where the commutator is

$$\begin{aligned}\left[\left[\tilde{D},i\sigma^iE^i\right],\phi\right] &= -\left[\phi,\tilde{D}i\sigma^iE^i\right]+\left[\phi,i\sigma^iE^i\tilde{D}\right]\\ &= -\left[\phi,\tilde{D}\right]i\sigma^iE^i-\tilde{D}\left[\phi,i\sigma^iE^i\right]+\left[\phi,i\sigma^iE^i\right]\tilde{D}+i\sigma^iE^i\left[\phi,\tilde{D}\right]\\ &= -\left[\phi,\tilde{D}\right]i\sigma^iE^i+i\sigma^iE^i\left[\phi,\tilde{D}\right]=\frac{\hbar}{2mc}\left(\sigma^j\sigma^i-\sigma^i\sigma^j\right)E^i(\partial_j\phi)\\ &= \frac{\hbar}{2mc}i2\epsilon^{jik}\sigma^kE^i(\partial_j\phi)=\frac{-i\hbar}{2mc}2\,\boldsymbol{\sigma}\cdot\mathbf{E}\times(\nabla\phi),\end{aligned} \tag{A.19}$$

so that

$$\begin{aligned}\Omega H_\alpha\Omega^{-1} &= -e\phi+\frac{(\mathbf{p}+e\mathbf{A})^2}{2m}+\frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}-\frac{1}{2mc^2}\left(\frac{(\mathbf{p}+e\mathbf{A})^2}{2m}+\frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)^2\\ &-e\left(\frac{-i\hbar}{2mc}\right)^2\sigma^j\sigma^iD_jE^i-e\left(\frac{-i\hbar}{2mc}\right)^3\sigma^j\sigma^iD_j(\partial_0E^i)\\ &-\left(\frac{-i\hbar}{2mc}\right)^2\frac{e}{2}\left(\partial_i^2\phi+2(\partial_i\phi)D_i\right)+\frac{ie^2\hbar^2}{(2mc)^3c}\boldsymbol{\sigma}\cdot\mathbf{E}\times(\nabla\phi).\end{aligned} \tag{A.20}$$

Putting the $\mathcal{O}(m^{-3})$ term of H_Ψ together, one obtains

$$-\frac{1}{2mc^2}\left(\frac{(\mathbf{p}+e\mathbf{A})^2}{2m}+\frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\mathbf{B}\right)^2-e\left(\frac{-i\hbar}{2mc}\right)^3\sigma^j\sigma^iD_j(\partial_0E^i)+$$

$$
\begin{aligned}
&+ \frac{ie^2\hbar^2}{(2mc)^3 c}\boldsymbol{\sigma}\cdot\mathbf{E}\times(\nabla\phi) + \frac{e}{2c}\left(\frac{-i\hbar}{2mc}\right)^3 \big(\sigma^i\sigma^j(\partial_t\partial_i E^j) - 2i\epsilon^{ijk}\sigma^k(\partial_t E^i D_j)\big) \\
&= \frac{1}{(2mc)^3}\Bigg[-c\big((\mathbf{p}+e\mathbf{A})^2 + e\hbar\boldsymbol{\sigma}\cdot\mathbf{B}\big)^2 + \frac{ie^2\hbar^2}{c}\boldsymbol{\sigma}\cdot\mathbf{E}\times(\nabla\phi) \\
&- \frac{e}{2c}(-i\hbar)^3\sigma^j\sigma^i\left\{(\partial_j\partial_t E^i) + (\partial_t E^i)D_j + D_j(\partial_t E^i)\right\} \\
&+ \frac{e(-i\hbar)^3}{2c}\left(\sigma^i\sigma^j(\partial_t\partial_i E^j) - 2i\epsilon^{ijk}\sigma^k\left\{(\partial_t E^i)D_j + \frac{ie}{\hbar}E^i(\partial_t A^j)\right\}\right)\Bigg] \\
&= \frac{1}{(2mc)^3}\Bigg[-c\big((\mathbf{p}+e\mathbf{A})^2 + e\hbar\boldsymbol{\sigma}\cdot\mathbf{B}\big)^2 + \frac{e(-i\hbar)^3}{2c}\big(-2i\epsilon^{ijk}\sigma^k(\partial_t E^i)D_j\big) \\
&- \frac{e}{2c}(-i\hbar)^3\sigma^j\sigma^i\left\{(\partial_t E^i)D_j + D_j(\partial_t E^i)\right\} - \frac{ie^2\hbar^2}{c}\boldsymbol{\sigma}\cdot\mathbf{E}\times\mathbf{E}\Bigg] \\
&= \frac{1}{(2mc)^3}\Bigg[-c\big((\mathbf{p}+e\mathbf{A})^2 + e\hbar\boldsymbol{\sigma}\cdot\mathbf{B}\big)^2 \\
&- \frac{e\,(-i\hbar)^3}{2c}\left\{(\partial_t E^i)D_i + D_i(\partial_t E^i) - i\epsilon^{ijk}\sigma^k(\partial_t\partial_j E^i)\right\}\Bigg], \qquad \text{(A.21)}
\end{aligned}
$$

giving Eq. (2.23).

Appendix B

$YTiO_3$

This appendix contains some of the lengthy expressions involved in calculating the RIXS spectra of $YTiO_3$ (section 5.4).

B.1 RIXS – Single site processes

The angular momentum $\hat{l}$ and quadrupole operators $\hat{Q}, \hat{T}$ in Eqs. (5.43–5.45) are defined as follows:

$$\hat{l}_x = i(c^\dagger b - b^\dagger c) \tag{B.1}$$

$$\hat{l}_y = i(a^\dagger c - c^\dagger a) \tag{B.2}$$

$$\hat{l}_z = i(b^\dagger a - a^\dagger b) \tag{B.3}$$

$$\hat{Q}_x = \hat{l}_x^2 - \hat{l}_y^2 = n_b - n_a \tag{B.4}$$

$$\hat{Q}_z = \frac{1}{\sqrt{3}}(\hat{l}_x^2 + \hat{l}_y^2 - 2\hat{l}_z^2) = \frac{1}{\sqrt{3}}(2n_c - n_a - n_b) \tag{B.5}$$

$$\hat{T}_x = \hat{l}_y\hat{l}_z + \hat{l}_z\hat{l}_y = -(b^\dagger c + c^\dagger b) \tag{B.6}$$

$$\hat{T}_y = \hat{l}_x\hat{l}_z + \hat{l}_z\hat{l}_x = -(c^\dagger a + a^\dagger c) \tag{B.7}$$

$$\hat{T}_z = \hat{l}_x\hat{l}_y + \hat{l}_y\hat{l}_x = -(a^\dagger b + b^\dagger a) \tag{B.8}$$

which are normalized by $\mathrm{Tr}\left(\hat{\Gamma}^2\right) = 2$. The corresponding matrices $\Gamma_{d'd}$ in Eq. (5.41) are

$$\Gamma^{Q_x} = \frac{1}{2}\begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \qquad \Gamma^{Q_z} = \frac{1}{2\sqrt{3}}\begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{pmatrix},$$
$$\Gamma^{T_x} = -\frac{1}{2}\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \qquad \Gamma^{T_y} = -\frac{1}{2}\begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix},$$
$$\Gamma^{T_z} = -\frac{1}{2}\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \qquad \Gamma^{l_x} = \frac{1}{2}\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \end{pmatrix},$$
$$\Gamma^{l_y} = \frac{1}{2}\begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, \qquad \Gamma^{l_z} = \frac{1}{2}\begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{B.9}$$

with the indices $d, d' = (yz, zx, xy)$ (or for polarization dependence: $\alpha, \beta = (x, y, z)$).

B.2 Multiplet factors

For the multiplet effect factors in Eq. (5.46), we have

$$M_{d'd}^{A_{1g}} = \sqrt{\frac{2}{3}}\left(\langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle + \langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle + \langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle\right) \tag{B.10}$$
$$M_{d'd}^{Q_x} = \left(\langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle - \langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle\right) \tag{B.11}$$
$$M_{d'd}^{Q_z} = \frac{1}{\sqrt{3}}\left(2\langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle - \langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle - \langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle\right) \tag{B.12}$$
$$M_{d'd}^{T_x} = -\left(\langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle + \langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle\right) \tag{B.13}$$
$$M_{d'd}^{T_y} = -\left(\langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle + \langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle\right) \tag{B.14}$$
$$M_{d'd}^{T_z} = -\left(\langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle + \langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle\right) \tag{B.15}$$
$$M_{d'd}^{l_x} = -i\left(\langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle - \langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle\right) \tag{B.16}$$
$$M_{d'd}^{l_y} = -i\left(\langle d'|\,\hat{z}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle - \langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{z}\,|d\rangle\right) \tag{B.17}$$
$$M_{d'd}^{l_z} = -i\left(\langle d'|\,\hat{x}\,|m\rangle\langle m|\,\hat{y}\,|d\rangle - \langle d'|\,\hat{y}\,|m\rangle\langle m|\,\hat{x}\,|d\rangle\right) \tag{B.18}$$

Note that the position operators act on the core electrons, not the t_{2g} ones. Both the core and t_{2g} electrons are implied in the states $|d\rangle, |d'\rangle$.

B.3 The operators $\hat{\Gamma}$ in terms of orbitons

In terms of the orbiton operators, we obtain the one-orbiton creation part of $\hat{\Gamma}_{\mathbf{q}} = \sum_i e^{i\mathbf{q}\cdot\mathbf{r}_i}\hat{\Gamma}_i$ to be

$$\begin{aligned}\hat{l}^{(1)}_{x,\mathbf{q}-\mathbf{q}_1} &= \frac{i\,|c_0|}{2}\sqrt{\frac{N}{3}}\Big[\Big\{(1-\sqrt{3})u_{\mathbf{q}} + (1+\sqrt{3})v_{\mathbf{q}}\Big\}(\text{sh }\theta_{1,\mathbf{q}} + \text{ch }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + \Big\{(-1-\sqrt{3})u_{\mathbf{q}} + (1-\sqrt{3})v_{\mathbf{q}}\Big\}(\text{sh }\theta_{2,\mathbf{q}} + \text{ch }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.19})\end{aligned}$$

$$\begin{aligned}\hat{l}^{(1)}_{y,\mathbf{q}-\mathbf{q}_2} &= \frac{i\,|c_0|}{2}\sqrt{\frac{N}{3}}\Big[\Big\{(1+\sqrt{3})u_{\mathbf{q}} + (1-\sqrt{3})v_{\mathbf{q}}\Big\}(\text{sh }\theta_{1,\mathbf{q}} + \text{ch }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + \Big\{(-1+\sqrt{3})u_{\mathbf{q}} + (1+\sqrt{3})v_{\mathbf{q}}\Big\}(\text{sh }\theta_{2,\mathbf{q}} + \text{ch }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.20})\end{aligned}$$

$$\begin{aligned}\hat{l}^{(1)}_{z,\mathbf{q}-\mathbf{q}_3} &= -i\,|c_0|\sqrt{\frac{N}{3}}\Big[(u_{\mathbf{q}} + v_{\mathbf{q}})(\text{sh }\theta_{1,\mathbf{q}} + \text{ch }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + (v_{\mathbf{q}} - u_{\mathbf{q}})(\text{sh }\theta_{2,\mathbf{q}} + \text{ch }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.21})\end{aligned}$$

$$\begin{aligned}\hat{T}^{(1)}_{x,\mathbf{q}-\mathbf{q}_1} &= \frac{|c_0|}{6}\sqrt{N}\Big[\Big\{(1+\sqrt{3})u_{\mathbf{q}} + (-1+\sqrt{3})v_{\mathbf{q}}\Big\}(\text{ch }\theta_{1,\mathbf{q}} - \text{sh }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + \Big\{(1-\sqrt{3})u_{\mathbf{q}} + (1+\sqrt{3})v_{\mathbf{q}}\Big\}(\text{ch }\theta_{2,\mathbf{q}} - \text{sh }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.22})\end{aligned}$$

$$\begin{aligned}\hat{T}^{(1)}_{y,\mathbf{q}-\mathbf{q}_2} &= \frac{|c_0|}{6}\sqrt{N}\Big[\Big\{(1-\sqrt{3})u_{\mathbf{q}} + (-1-\sqrt{3})v_{\mathbf{q}}\Big\}(\text{ch }\theta_{1,\mathbf{q}} - \text{sh }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + \Big\{(1+\sqrt{3})u_{\mathbf{q}} + (1-\sqrt{3})v_{\mathbf{q}}\Big\}(\text{ch }\theta_{2,\mathbf{q}} - \text{sh }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.23})\end{aligned}$$

$$\begin{aligned}\hat{T}^{(1)}_{z,\mathbf{q}-\mathbf{q}_3} &= -\frac{|c_0|}{3}\sqrt{N}\Big[(u_{\mathbf{q}} - v_{\mathbf{q}})(\text{ch }\theta_{1,\mathbf{q}} - \text{sh }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + (u_{\mathbf{q}} + v_{\mathbf{q}})(\text{ch }\theta_{2,\mathbf{q}} - \text{sh }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.24})\end{aligned}$$

$$\begin{aligned}\hat{Q}^{(1)}_{x,\mathbf{q}} &= |c_0|\sqrt{\frac{N}{3}}\Big[-(u_{\mathbf{q}} + v_{\mathbf{q}})(\text{ch }\theta_{1,\mathbf{q}} - \text{sh }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + (u_{\mathbf{q}} - v_{\mathbf{q}})(\text{ch }\theta_{2,\mathbf{q}} - \text{sh }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.25})\end{aligned}$$

$$\begin{aligned}\hat{Q}^{(1)}_{z,\mathbf{q}} &= -\,|c_0|\sqrt{\frac{N}{3}}\Big[(u_{\mathbf{q}} - v_{\mathbf{q}})(\text{ch }\theta_{1,\mathbf{q}} - \text{sh }\theta_{1,\mathbf{q}})\alpha^\dagger_{1,-\mathbf{q}} \\ &\quad + (u_{\mathbf{q}} + v_{\mathbf{q}})(\text{ch }\theta_{2,\mathbf{q}} - \text{sh }\theta_{2,\mathbf{q}})\alpha^\dagger_{2,-\mathbf{q}}\Big] \qquad (\text{B.26})\end{aligned}$$

with $\mathbf{q}_1 = (\pi, 0, \pi)$, $\mathbf{q}_2 = (\pi, \pi, 0)$, $\mathbf{q}_3 = (0, \pi, \pi)$. The expressions for the two-orbiton creation part of $\hat{\Gamma}_{\mathbf{q}} = \sum_i e^{i\mathbf{q}\cdot\mathbf{r}_i}\hat{\Gamma}_i$ are

$$\hat{l}^{(2)}_{x,\mathbf{q}} = \frac{i}{\sqrt{3}}\sum_{\mathbf{k}}\Big[(vu' - uv')\text{ch }\theta_1\text{ sh }\theta_1'\;\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}} +$$

$$+ (vu' - uv')\text{ch}\,\theta_2\ \text{sh}\,\theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}}$$
$$+ (uu' + vv')(\text{ch}\,\theta_1\ \text{sh}\,\theta_2' - \text{sh}\,\theta_1\ \text{ch}\,\theta_2')\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}}\Big] \tag{B.27}$$

with $u, v, \theta_1, \theta_2 = u_\mathbf{k}, v_\mathbf{k}, \theta_{1,\mathbf{k}}, \theta_{2,\mathbf{k}}$ and primed quantities $u', v', \theta_1', \theta_2' = u_{\mathbf{k}+\mathbf{q}_1+\mathbf{q}}$, $v_{\mathbf{k}+\mathbf{q}_1+\mathbf{q}}$, $\theta_{1,\mathbf{k}+\mathbf{q}_1+\mathbf{q}}$, $\theta_{2,\mathbf{k}+\mathbf{q}_1+\mathbf{q}}$. Further, $\hat{l}^{(2)}_{y,\mathbf{q}}$ and $\hat{l}^{(2)}_{z,\mathbf{q}}$ have the same form as $\hat{l}^{(2)}_{x,\mathbf{q}}$ but with $\mathbf{q}_1$ replaced by $\mathbf{q}_2$ and $\mathbf{q}_3$ respectively. Next,

$$\hat{T}^{(2)}_{x,\mathbf{q}} = \sum_\mathbf{k}\Bigg[\left\{-(uu'+vv') + \frac{uu'-vv'}{\sqrt{3}} + \frac{uv'+vu'}{3}\right\}\text{ch}\,\theta_1\ \text{sh}\,\theta_1'\ \alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}}$$
$$+\left\{-(uu'+vv') - \frac{uu'-vv'}{\sqrt{3}} - \frac{uv'+vu'}{3}\right\}\text{ch}\,\theta_2\ \text{sh}\,\theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}}$$
$$+\left\{-(uv'-vu') - \frac{uv'+vu'}{\sqrt{3}} - \frac{uu'-vv'}{3}\right\}(\text{ch}\,\theta_1\ \text{sh}\,\theta_2' + \text{sh}\,\theta_1\ \text{ch}\,\theta_2')\times$$
$$\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_1-\mathbf{q}}\Bigg] \tag{B.28}$$

$$\hat{T}^{(2)}_{y,\mathbf{q}} = \sum_\mathbf{k}\Bigg[\left\{-(uu'+vv') - \frac{uu'-vv'}{\sqrt{3}} + \frac{uv'+vu'}{3}\right\}\text{ch}\,\theta_1\ \text{sh}\,\theta_1'\ \alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}_2-\mathbf{q}}$$
$$+\left\{-(uu'+vv') + \frac{uu'-vv'}{\sqrt{3}} - \frac{uv'+vu'}{3}\right\}\text{ch}\,\theta_2\ \text{sh}\,\theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_2-\mathbf{q}}$$
$$+\left\{-(uv'-vu') + \frac{uv'+vu'}{\sqrt{3}} - \frac{uu'-vv'}{3}\right\}(\text{ch}\,\theta_1\ \text{sh}\,\theta_2' + \text{sh}\,\theta_1\ \text{ch}\,\theta_2')\times$$
$$\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_2-\mathbf{q}}\Bigg] \tag{B.29}$$

where in the expression for $\hat{T}^{(2)}_{y,\mathbf{q}}$ we replaced $\mathbf{q}_1$ by $\mathbf{q}_2$: $u', v', \theta_1', \theta_2' = u_{\mathbf{k}+\mathbf{q}_2+\mathbf{q}}$, $v_{\mathbf{k}+\mathbf{q}_2+\mathbf{q}}, \theta_{1,\mathbf{k}+\mathbf{q}_2+\mathbf{q}}, \theta_{2,\mathbf{k}+\mathbf{q}_2+\mathbf{q}}$.

$$\hat{T}^{(2)}_{z,\mathbf{q}} = \sum_\mathbf{k}\Bigg[\left\{-\frac{2}{3}(uv'+vu') - (uu'+vv')\right\}\text{ch}\,\theta_1\ \text{sh}\,\theta_1'\ \alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}_3-\mathbf{q}}$$
$$+\left\{\frac{2}{3}(uv'+vu') - (uu'+vv')\right\}\text{ch}\,\theta_2\ \text{sh}\,\theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_3-\mathbf{q}}$$
$$+\left\{\frac{2}{3}(uu'-vv') - (uv'-vu')\right\}(\text{ch}\,\theta_1\ \text{sh}\,\theta_2' + \text{sh}\,\theta_1\ \text{ch}\,\theta_2')\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}_3-\mathbf{q}}\Bigg] \tag{B.30}$$

where we replaced $\mathbf{q}_1$ by $\mathbf{q}_3$: $u', v', \theta_1', \theta_2' = u_{\mathbf{k}+\mathbf{q}_3+\mathbf{q}}$, $v_{\mathbf{k}+\mathbf{q}_3+\mathbf{q}}$, $\theta_{1,\mathbf{k}+\mathbf{q}_3+\mathbf{q}}$, $\theta_{2,\mathbf{k}+\mathbf{q}_3+\mathbf{q}}$. Finally,

$$\hat{Q}^{(2)}_{x,\mathbf{q}} = -\frac{1}{\sqrt{3}}\sum_\mathbf{k}\Big[-(uu'-vv')\text{ch}\,\theta_1\ \text{sh}\,\theta_1'\ \alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}}$$
$$+ (uu'-vv')\text{ch}\,\theta_2\ \text{sh}\,\theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}} +$$

$$+ (uv' + vu')(\text{ch}\ \theta_1\ \text{sh}\ \theta_2' + \text{sh}\ \theta_1\ \text{ch}\ \theta_2')\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}}\Big] \tag{B.31}$$

$$\hat{Q}^{(2)}_{z,\mathbf{q}} = \frac{1}{\sqrt{3}}\sum_{\mathbf{k}}\Big[(uv' + vu')\text{ch}\ \theta_1\ \text{sh}\ \theta_1'\ \alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{1,-\mathbf{k}-\mathbf{q}}$$
$$- (uv' + vu')\text{ch}\ \theta_2\ \text{sh}\ \theta_2'\ \alpha^\dagger_{2,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}}$$
$$- (uu' - vv')(\text{ch}\ \theta_1\ \text{sh}\ \theta_2' + \text{sh}\ \theta_1\ \text{ch}\ \theta_2')\alpha^\dagger_{1,\mathbf{k}}\alpha^\dagger_{2,-\mathbf{k}-\mathbf{q}}\Big] \tag{B.32}$$

where in both equations we replaced $\mathbf{q}_1$ by $\mathbf{0}$: $u', v', \theta_1', \theta_2' = u_{\mathbf{k}+\mathbf{q}}$, $v_{\mathbf{k}+\mathbf{q}}$, $\theta_{1,\mathbf{k}+\mathbf{q}}$, $\theta_{2,\mathbf{k}+\mathbf{q}}$.

B.4 RIXS – two-site processes with superexchange model

Functions f_{11}, f_{22} and f_{12} in Eqs. (5.79–5.80) are:

$$f_{11}(\mathbf{k},\mathbf{q}) = [-\gamma_{3,\mathbf{q}}(uv' + u'v) - \gamma_{2,\mathbf{q}}(uu' - vv') - (1+\gamma_{1,\mathbf{q}})(uu' + vv')] \times$$
$$(\text{ch}\ \theta_1\ \text{sh}\ \theta_1' + \text{sh}\ \theta_1\ \text{ch}\ \theta_1')$$
$$+ 2[\gamma_1'(uu' + vv') + \gamma_2'(uu' - vv') + \gamma_3'(uv' + u'v)] \times$$
$$(\text{sh}\ \theta_1\ \text{sh}\ \theta_1' + \text{ch}\ \theta_1\ \text{ch}\ \theta_1') \tag{B.33}$$

$$f_{22}(\mathbf{k},\mathbf{q}) = [\gamma_{3,\mathbf{q}}(uv' + u'v) + \gamma_{2,\mathbf{q}}(uu' - vv') - (1+\gamma_{1,\mathbf{q}})(uu' + vv')] \times$$
$$(\text{ch}\ \theta_2\ \text{sh}\ \theta_2' + \text{sh}\ \theta_2\ \text{ch}\ \theta_2')$$
$$+ 2[\gamma_1'(uu' + vv') - \gamma_2'(uu' - vv') - \gamma_3'(uv' + u'v)] \times$$
$$(\text{sh}\ \theta_2\ \text{sh}\ \theta_2' + \text{ch}\ \theta_2\ \text{ch}\ \theta_2') \tag{B.34}$$

$$f_{12}(\mathbf{k},\mathbf{q}) = 2[\gamma_{3,\mathbf{q}}(uu' - vv') + \gamma_{2,\mathbf{q}}(uv' + u'v) - (1+\gamma_{1,\mathbf{q}})(uv' - u'v)] \times$$
$$(\text{ch}\ \theta_1\ \text{sh}\ \theta_2' + \text{sh}\ \theta_1\ \text{ch}\ \theta_2')$$
$$+ 4[\gamma_1'(uv' - u'v) + \gamma_2'(uv' + u'v) - \gamma_3'(uu' - vv')] \times$$
$$(\text{sh}\ \theta_1\ \text{sh}\ \theta_2' + \text{ch}\ \theta_1\ \text{ch}\ \theta_2') \tag{B.35}$$

where we shortened notation by writing $\theta_{1/2} = \theta_{1/2,\mathbf{k}}$, $\theta'_{1/2} = \theta_{1/2,\mathbf{k}+\mathbf{q}}$, $u(') = u_{\mathbf{k}(+\mathbf{q})}$, $v(') = v_{\mathbf{k}(+\mathbf{q})}$ and $\gamma_i' = \gamma_{i,\mathbf{k}+\mathbf{q}}$.

Appendix C

Phonon RIXS

This appendix contains the lengthy derivations involved in calculating the Einstein phonon RIXS spectra of chapter 7. In the following, we evaluate the scattering amplitude (7.8). We use (see Mahan [62], section 4.3.2)

$$
\begin{aligned}
\langle n| e^{\frac{M}{\omega_0}(b^\dagger - b)} \left| n^0 \right\rangle &= e^{-g/2} \langle n| e^{\frac{M}{\omega_0} b^\dagger} e^{-\frac{M}{\omega_0} b} \left| n^0 \right\rangle \\
&= e^{-g/2} \sum_{k=0}^{n} \sum_{l=0}^{n^0} \langle n-k| \frac{(M/\omega_0)^k}{k!} \left[\frac{n!}{(n-k)!} \right]^{\frac{1}{2}} \frac{(-M/\omega_0)^l}{l!} \left[\frac{n^0!}{(n^0-l)!} \right]^{\frac{1}{2}} \left| n^0 - l \right\rangle \\
&= e^{-g/2} \sum_{k=0}^{n} \sum_{l=0}^{n^0} \frac{(M/\omega_0)^k}{k!} \left[\frac{n! n^0!}{(n-k)!(n^0-l)!} \right]^{\frac{1}{2}} \frac{(-M/\omega_0)^l}{l!} \delta_{n^0-l, n-k} \\
&= \begin{cases} e^{-g/2} \sum_{l=0}^{n^0} \frac{(M/\omega_0)^{l-n^0+n}}{(l-n^0+n)!} \left[\frac{n! n^0!}{(n-(l-n^0+n))!(n^0-l)!} \right]^{\frac{1}{2}} \frac{(-M/\omega_0)^l}{l!} & \text{for } n > n^0 \\ e^{-g/2} \sum_{k=0}^{n} \frac{(M/\omega_0)^k}{k!} \left[\frac{n! n^0!}{(n-k)!(n^0-(n^0-n+k))!} \right]^{\frac{1}{2}} \frac{(-M/\omega_0)^{n^0-n+k}}{(n^0-n+k)!} & \text{for } n \leq n^0 \end{cases} \\
&= \begin{cases} e^{-g/2} \sum_{l=0}^{n^0} \frac{(-1)^l (M/\omega_0)^{2l-n^0+n}}{l!(l-n^0+n)!} \frac{\sqrt{n! n^0!}}{(n^0-l)!} & \text{for } n > n^0 \\ e^{-g/2} \sum_{l=0}^{n} \frac{(-1)^{l+n^0-n} (M/\omega_0)^{2l+n^0-n}}{l!(l+n^0-n)!} \frac{\sqrt{n! n^0!}}{(n-l)!} & \text{for } n \leq n^0 \end{cases}
\end{aligned} \tag{C.1}
$$

with $g = m^2/\omega_0^2$. For simplicity, we assume the system is initially in its ground state: $n^0 = 0$. In that case, the above expression simplifies to

$$
\langle n| e^{\frac{M}{\omega_0}(b^\dagger - b)} |0\rangle = e^{-g/2} \frac{(M/\omega_0)^n}{\sqrt{n!}}. \tag{C.2}
$$

These expressions are inserted in the Kramers-Heisenberg equation (7.12):

$$\mathcal{F}_{fg} = T_{\mathrm{el}}(\boldsymbol{\epsilon}', \boldsymbol{\epsilon}) \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} F_i \tag{C.3}$$

with

$$\begin{aligned} F_i = {} & e^{-g} \sum_{n=0}^{n_i'} \sum_{l=0}^{n} \frac{(-1)^{l+n_i'-n}\,(M/\omega_0)^{2l+n_i'}}{l!(l+n_i'-n)!} \frac{\sqrt{n_i'!}}{(n-l)!} \frac{1}{z+(g-n)\omega_0} \\ & + e^{-g} \sum_{n=n_i'+1}^{\infty} \sum_{l=0}^{n_i'} \frac{(-1)^{l}\,(M/\omega_0)^{2l-n_i'+2n}}{l!(l-n_i'+n)!} \frac{\sqrt{n_i'!}}{(n_i'-l)!} \frac{1}{z+(g-n)\omega_0}. \end{aligned} \tag{C.4}$$

Appendix D

Magnetic spectral weight at the Γ point in 2D cuprates

This appendix contains the calculational details from Sec. 4.5.5, where the leading magnetic contribution to the $\mathbf{q} = \mathbf{0}$ RIXS spectra at the Cu L edge is established.

D.1 $\bar{H}_{\text{eff}}^{(4)}$ to fourth order in t/U

In this section we evaluate Eq. (4.74) in the presence of a core hole.

The first term of Eq. (4.74) changes the Hamiltonian to

$$\bar{H}_{\text{eff}}^{(4)} = H_0 - \frac{4t^2}{U} \sum_i p_i p_i^\dagger \sum_\delta \mathbf{S}_i \cdot \mathbf{S}_{i+\delta} + \dots \tag{D.1}$$

where $p_i^\dagger$ creates a core electron, δ points to nearest neighbors, and the dots indicate the corrections to H_0 due to the four hop terms.

To handle the four hop terms systematically, we categorize all terms according to the 'connections' between sites. With a 'connection' is meant that one or more hops occur between the sites in question. For example, only the sites i and j, and j and k are connected if we select the following four hoppings from the V's:

$$t_{ij} \sum_\sigma (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) \times \dots \times t_{jk} \sum_{\sigma'} (c_{j\sigma'}^\dagger c_{k\sigma'} + c_{j\sigma'}^\dagger c_{k\sigma'}) \times \dots \times t_{ij}(\dots) \times \dots \times t_{jk}(\dots), \tag{D.2}$$

but also processes with a different hopping order like $t_{ij}t_{ij}t_{jk}t_{jk}$ and $t_{jk}t_{ij}t_{ij}t_{jk}$, etc., have the same connections and are thus categorized together. We will therefore represent the category comprising all these processes by $t_{ij}^2 t_{jk}^2$. The order of the t's is not important, only the number of times a certain hop occurs is important for indicating the category.

We now establish the different categories. The most general string of t's is $t_{ij}t_{kl}t_{mn}t_{pq}$. A lot of these processes change the occupancy of one of the sites. These terms vanish because of the P_0's at the beginning and end of every four hop term. This imposes a restriction on the indices i, j, k, l, m, n, p, q: if an index appears an odd number of times, the occupancy is changed and the process does not contribute to $H_{\text{eff}}^{(4)}$. We are left with the following categories: **(a)** t_{ij}^4, **(b)** $t_{ij}^2 t_{kl}^2 (i, j \neq k, l)$, **(c)** $t_{ij}^2 t_{jk}^2 (i \neq k)$, and **(d)** $t_{ij}t_{jk}t_{kl}t_{li}$ (i, j, k, l form a square). If there is no core hole present at any of the sites involved, we can just copy-paste the results from Ref. [82]. Below we analyze the processes where there is a core hole present.

Processes in category (**a**) do not flip any spins. The doublon (the doubly occupied site) hops 4 times between site i and j, where i is the core hole site. We obtain the (**a**) contribution:

$$H_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \frac{t^4}{U_c^3} P_0 \sum_\sigma c_{i\sigma}^\dagger c_{j\sigma} c_{j\sigma}^\dagger c_{i\sigma} P_0 \, c_{i\sigma}^\dagger c_{j\sigma} c_{j\sigma}^\dagger c_{i\sigma} P_0 \,, \tag{D.3}$$

where the dots indicate other fourth order terms. At the core hole site, $c_{i\sigma}^\dagger c_{i\sigma} = 1$, and we use the projected spins as defined in Eq. (2.14) of Ref. [82] to get

$$H_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \frac{t^4}{U_c^3} \left(\{\frac{1}{2}(1 - \sigma_j^z)\}^2 + \{\frac{1}{2}(1 + \sigma_j^z)\}^2 \right) = \cdots + p_i p_i^\dagger \frac{t^4}{U_c^3} \mathbb{1}. \tag{D.4}$$

The corresponding part of H_0 is

$$H_0 = \cdots - \frac{16t^4}{U^3} \mathbf{S}_i \cdot \mathbf{S}_j \tag{D.5}$$

which should be replaced by Eq. (D.4). Dropping the constant term, this gives

$$\bar{H}_{\text{eff}}^{(4)} = H_0 + \sum_i p_i p_i^\dagger \sum_\delta \frac{16t^4}{U^3} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta} + \ldots \tag{D.6}$$

Processes in category **(b)** do not appear in the fourth order expansion of the half-filled Hubbard model. If we add one doubly occupied site (namely, the core hole site i), there still is no contribution to $\bar{H}_{\text{eff}}^{(4)}$. The only matrix elements for which this is not a priori clear, involve configurations pictured in Fig. D.1.

If the spins at k and l are parallel, the corresponding matrix element is obviously zero. Working out the other matrix elements (12 pathways for interchanging anti-parallel k and l, and 12 pathways for leaving anti-parallel k and l invariant),

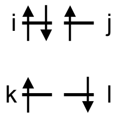

Figure D.1: The processes of category **(b)** involve $t_{ij}^2 t_{kl}^2$, connecting i to j and k to l.

Figure D.2: The processes of category **(c)** are subdivided into two cases: the core hole site is connected to one other site (left), or the core hole site is connected to two other sites (right).

it turns out that all pathways interfere to give 0, just as in the half-filled case. Therefore, H_0 is not modified by category **(b)** processes.

Processes in category **(c)** connect three sites i, j and k. They can be subdivided into two cases: in the first one, the core hole site i is connected to one other site, and the second one, the core hole site i is connected to two other sites. Starting with the first case, we have for the following matrix elements:

$$\uparrow_j\uparrow_k\to\uparrow_j\uparrow_k \text{ and } \downarrow_j\downarrow_k\to\downarrow_j\downarrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \frac{t^4}{U_c^3}\left(\frac{1}{2}\mathbb{1} + 2S_j^z S_k^z\right), \tag{D.7}$$

$$\uparrow_j\downarrow_k\to\uparrow_j\downarrow_k \text{ and } \downarrow_j\uparrow_k\to\downarrow_j\uparrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \frac{t^4}{U_c^3}\left(\frac{1}{2}\mathbb{1} - 2S_j^z S_k^z\right), \tag{D.8}$$

$$\uparrow_j\downarrow_k\to\downarrow_j\uparrow_k \text{ and } \downarrow_j\uparrow_k\to\uparrow_j\downarrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots + 0. \tag{D.9}$$

Adding these contributions and dropping the constants yields

$$\bar{H}_{\text{eff}}^{(4)} = \cdots + 0. \tag{D.10}$$

The second case gives

$$\uparrow_j\uparrow_k\to\uparrow_j\uparrow_k \text{ and } \downarrow_j\downarrow_k\to\downarrow_j\downarrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \frac{2t^4}{U_c^3}\left(\frac{1}{2}\mathbb{1} + 2S_j^z S_k^z\right), \tag{D.11}$$

$$\uparrow_j\downarrow_k\to\uparrow_j\downarrow_k \text{ and } \downarrow_j\uparrow_k\to\downarrow_j\uparrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \left(\frac{2t^4}{UU_c^2} + \frac{4t^4}{U_c^2(2U_c+U)} - \frac{2t^4}{U_c^3}\right)\left(\frac{1}{2}\mathbb{1} - 2S_j^z S_k^z\right), \tag{D.12}$$

$$\uparrow_j\downarrow_k\to\downarrow_j\uparrow_k \text{ and } \downarrow_j\uparrow_k\to\uparrow_j\downarrow_k : \quad \bar{H}_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \left(\frac{2t^4}{UU_c^2} + \frac{4t^4}{U_c^2(2U_c+U)}\right) \times \left(S_j^+ S_k^- + S_j^- S_k^+\right). \tag{D.13}$$

Adding the contributions and dropping the constants yields

$$\bar{H}_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \left(\frac{4t^4}{UU_c^2} + \frac{8t^4}{U_c^2(2U_c+U)}\right) \mathbf{S}_j \cdot \mathbf{S}_k. \tag{D.14}$$

The corresponding part of H_0 (including the second neighbor terms from category (**d**))[1] is

$$H_0 = \cdots + \sum_{j \neq k} \frac{4t_{ij}^2 t_{ik}^2}{U^3} \mathbf{S}_j \cdot \mathbf{S}_k \tag{D.15}$$

and it is replaced by

$$\bar{H}_{\text{eff}}^{(4)} = H_0 + \sum_i p_i p_i^\dagger \sum_{j \neq k} \left(\frac{4t^4}{UU_c^2} + \frac{8t^4}{U_c^2(2U_c + U)} - \frac{4t^4}{U^3} \right) \mathbf{S}_j \cdot \mathbf{S}_k + \ldots \tag{D.16}$$

where the sum over $j \neq k$ is over all pairs of neighbors j, k of i.

Finally, the processes of category **(d)** are

$$\begin{matrix} \uparrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} \rightarrow \begin{matrix} \uparrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} + \text{flipped}:$$

$$\bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \frac{2t^4}{U_c^3} \left(\frac{1}{4}\mathbb{1} + S_j^z S_k^z + S_k^z S_l^z + S_j^z S_l^z \right), \tag{D.17}$$

$$\begin{matrix} \uparrow_j & d_i \\ \downarrow_k & \uparrow_l \end{matrix} \rightarrow \begin{matrix} \uparrow_j & d_i \\ \downarrow_k & \uparrow_l \end{matrix} + \text{flipped}:$$

$$\bar{H}_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \frac{2t^4}{UU_c^2} \left(\frac{1}{4}\mathbb{1} - S_j^z S_k^z - S_k^z S_l^z + S_j^z S_l^z \right), \tag{D.18}$$

$$\begin{matrix} \downarrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} \rightarrow \begin{matrix} \downarrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} + \text{flip} + j \leftrightarrow l:$$

$$\bar{H}_{\text{eff}}^{(4)} = \cdots + p_i p_i^\dagger \frac{2t^4}{UU_c^2} \left(\frac{1}{2}\mathbb{1} - 2S_j^z S_l^z \right), \tag{D.19}$$

$$\begin{matrix} \downarrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} \rightarrow \begin{matrix} \uparrow_j & d_i \\ \downarrow_k & \uparrow_l \end{matrix} + \text{flip} + j \leftrightarrow l:$$

$$\begin{aligned} \bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \left(\frac{t^4}{U_c^3} + \frac{t^4}{UU_c^2} \right) \big(S_j^+ S_k^- + S_j^- S_k^+ \\ + S_k^+ S_l^- + S_k^- S_l^+ \big), \end{aligned} \tag{D.20}$$

$$\begin{matrix} \downarrow_j & d_i \\ \uparrow_k & \uparrow_l \end{matrix} \rightarrow \begin{matrix} \uparrow_j & d_i \\ \uparrow_k & \downarrow_l \end{matrix} + \text{flip} + j \leftrightarrow l:$$

$$\bar{H}_{\text{eff}}^{(4)} = \cdots - p_i p_i^\dagger \left(\frac{t^4}{U_c^3} + \frac{t^4}{UU_c^2} \right) \left(S_j^+ S_l^- + S_j^- S_l^+ \right). \tag{D.21}$$

where we have labeled the sites as shown in Fig. D.3, with d_i the doubly occupied core hole site.

[1] We do this because, first, it simplifies notation: the second and third neighbor terms in the sum get the same prefactor. Second, Coldea also does this, and following him makes it easy to compare to his mean field result for the ring exchange terms.

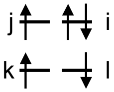

Figure D.3: The processes of category **(d)** ($t_{ij}t_{jk}t_{kl}t_{li}$) describe ring exchange.

Dropping constants, the final result for processes of category **(d)** is

$$H_{\text{eff}}^{(4)} = \cdots - \sum_{i,\text{squares at } i} p_i p_i^\dagger \left(\frac{2t^4}{UU_c^2} + \frac{2t^4}{U_c^3} \right) (\mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l + \mathbf{S}_j \cdot \mathbf{S}_l). \quad \text{(D.22)}$$

The corresponding terms in H_0 (minus the second neighbor terms, which were absorbed in the correction to H_0 due to category (**c**)) are

$$H_0 = \cdots - \frac{8t^4}{U^3} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \frac{80t^4}{U^3} \sum_{\text{squares}} [(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_i \cdot \mathbf{S}_l)(\mathbf{S}_j \cdot \mathbf{S}_k) - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l)]. \quad \text{(D.23)}$$

The mean field result is easily obtained [82, 83]:

$$H_0 = \cdots - \frac{48t^4}{U^3} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{20t^4}{U^3} \sum_{i,k} \mathbf{S}_i \cdot \mathbf{S}_k, \quad \text{(D.24)}$$

where the sum over i, k is over all pairs of next nearest neighbors. Then, at the mean field, the intermediate state Hamiltonian for processes of category (**d**) becomes

$$\begin{aligned} \bar{H}_{\text{eff}}^{(4)} = H_0 + \sum_i p_i p_i^\dagger \sum_{\text{squares at } i} \Bigg[& \frac{24t^4}{U^3} (\mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l + \mathbf{S}_l \cdot \mathbf{S}_i) \\ & + \frac{20t^4}{U^3} (\mathbf{S}_i \cdot \mathbf{S}_k + \mathbf{S}_j \cdot \mathbf{S}_l) \\ & - \left(\frac{2t^4}{UU_c^2} + \frac{2t^4}{U_c^3} \right) (\mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l + \mathbf{S}_j \cdot \mathbf{S}_l) \Bigg] + \ldots \end{aligned} \quad \text{(D.25)}$$

Adding all contributions from all categories, we obtain

$$\begin{aligned} \bar{H}_{\text{eff}}^{(4)} = H_0 + \sum_i p_i p_i^\dagger \Bigg\{ & \sum_j \left(\frac{16t^4}{U^3} - \frac{4t^2}{U} \right) \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{j \neq k} \Bigg(\frac{4t^4}{UU_c^2} + \frac{8t^4}{U_c^2(2U_c + U)} \\ & - \frac{4t^4}{U^3} \Bigg) \mathbf{S}_j \cdot \mathbf{S}_k + \sum_{\text{squares}} \Bigg[\frac{24t^4}{U^3} (\mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l + \mathbf{S}_l \cdot \mathbf{S}_i) \\ & + \frac{20t^4}{U^3} (\mathbf{S}_i \cdot \mathbf{S}_k + \mathbf{S}_j \cdot \mathbf{S}_l) - \left(\frac{2t^4}{UU_c^2} + \frac{2t^4}{U_c^3} \right) (\mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l \\ & + \mathbf{S}_j \cdot \mathbf{S}_l) \Bigg] \Bigg\}. \end{aligned} \quad \text{(D.26)}$$

The sums between the curly brackets are, respectively, over all nearest neighbors j of i, over all pairs of nearest neighbors j, k of i, and over all squares of 2×2 sites containing i. Grouping the different neighbor interactions, we finally get Eq. (4.75).

D.2 Scattering amplitude to fourth order in t/U

Once the intermediate state Hamiltonian is obtained to fourth order in t/U, it is straightforward and tedious to derive the RIXS scattering amplitude to this order. Below, we go through this expression term by term, where each term groups the interactions between certain neighbors.

The nearest neighbors term gives

$$\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} \sum_j^{\text{nn}} \mathbf{S}_i \cdot \mathbf{S}_j \left|0\right\rangle = \sum_{\mathbf{k}} \left[-\left(1+\gamma_{\mathbf{q}}\right)\left(U_{\mathbf{k}-\mathbf{q}}V_{\mathbf{k}} + U_{\mathbf{k}}V_{\mathbf{k}-\mathbf{q}}\right) + \left(\gamma_{\mathbf{k}-\mathbf{q}}+\gamma_{\mathbf{k}}\right)\left(U_{\mathbf{k}-\mathbf{q}}U_{\mathbf{k}} + V_{\mathbf{k}-\mathbf{q}}V_{\mathbf{k}}\right)\right] \alpha^\dagger_{\mathbf{k}}\alpha^\dagger_{-\mathbf{k}+\mathbf{q}}\left|0\right\rangle \tag{D.27}$$

as before.

For the next nearest neighbor term, we find

$$\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} \sum_j^{\text{nnn}} \mathbf{S}_i \cdot \mathbf{S}_j \left|0\right\rangle = -\sum_{\mathbf{k}} f_{\text{nnn}}(\mathbf{k},\mathbf{q})\left(U_{\mathbf{k}-\mathbf{q}}V_{\mathbf{k}} + U_{\mathbf{k}}V_{\mathbf{k}-\mathbf{q}}\right)\alpha^\dagger_{\mathbf{k}}\alpha^\dagger_{-\mathbf{k}+\mathbf{q}}\left|0\right\rangle . \tag{D.28}$$

where

$$f_{\text{nnn}}(\mathbf{k},\mathbf{q}) = \cos(k_x - q_x)\cos(k_y - q_y) + \cos k_x \cos k_y - 1 - \cos q_x \cos q_y. \tag{D.29}$$

The next term (with $j \neq k$ nearest neighbors of i) is

$$\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} \sum_{j\neq k} \mathbf{S}_j \cdot \mathbf{S}_k \left|0\right\rangle = -\sum_{\mathbf{k}} f_a(\mathbf{k},\mathbf{q})(U_{\mathbf{k}}V_{\mathbf{k}-\mathbf{q}} + U_{\mathbf{k}-\mathbf{q}}V_{\mathbf{k}})\alpha^\dagger_{\mathbf{k}}\alpha^\dagger_{-\mathbf{k}+\mathbf{q}}\left|0\right\rangle , \tag{D.30}$$

with

$$f_a(\mathbf{k},\mathbf{q}) = f_a(-\mathbf{k}-\mathbf{q},\mathbf{q}) = 2\gamma_{2\mathbf{k}-\mathbf{q}} - 6\gamma_{\mathbf{q}} + 2\cos k_x \cos(k_y - q_y) + 2\cos k_y \cos(k_x - q_x). \tag{D.31}$$

In the sum over squares, we have for the $\mathbf{S}_j \cdot \mathbf{S}_l$ term in the square sum:

$$\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} \sum_{\delta,\delta'} \mathbf{S}_{i+\delta} \cdot \mathbf{S}_{i+\delta'} \left|0\right\rangle = -\sum_{\mathbf{k}}(U_{\mathbf{k}}V_{\mathbf{k}-\mathbf{q}} + U_{\mathbf{k}-\mathbf{q}}V_{\mathbf{k}})f_b(\mathbf{k},\mathbf{q})\alpha^\dagger_{\mathbf{k}}\alpha^\dagger_{-\mathbf{k}+\mathbf{q}}\left|0\right\rangle \tag{D.32}$$

where the sum over δ, δ' is over the 4 pairs of orthogonal vectors that point to nearest neighbors of site i (they indicate $\mathbf{S}_j$ and $\mathbf{S}_l$), and

$$f_b(\mathbf{k},\mathbf{q}) = 2\left[-2\gamma_{\mathbf{q}} + \cos k_x \cos(k_y - q_y) + \cos k_y \cos(k_x - q_x)\right]. \tag{D.33}$$

For the square terms $\mathbf{S}_j \cdot \mathbf{S}_k + \mathbf{S}_k \cdot \mathbf{S}_l$ we find

$$\sum_{i,j,k} e^{i\mathbf{q}\cdot\mathbf{R}_i} \mathbf{S}_j \cdot \mathbf{S}_k \left|0\right\rangle = \sum_{\mathbf{k}} \Big(-(U_{\mathbf{k}-\mathbf{q}} V_{\mathbf{k}} + U_{\mathbf{k}} V_{\mathbf{k}-\mathbf{q}}) f_{c1}(\mathbf{q}) \\ + (U_{\mathbf{k}} U_{\mathbf{k}-\mathbf{q}} + V_{\mathbf{k}} V_{\mathbf{k}-\mathbf{q}}) f_{c2}(\mathbf{k},\mathbf{q}) \Big) \alpha^\dagger_{\mathbf{k}} \alpha^\dagger_{-\mathbf{k}+\mathbf{q}} \left|0\right\rangle \tag{D.34}$$

with j pointing to nearest neighbors of i, k to next nearest neighbors of i that are also nearest neighbors of j, and

$$f_{c1}(\mathbf{q}) = 2\gamma_{\mathbf{q}} + 2\cos q_x \cos q_y, \tag{D.35}$$

$$f_{c2}(\mathbf{k},\mathbf{q}) = \cos k_x \cos q_y + \cos k_y \cos q_x \\ + \cos(k_x - q_x)\cos q_y + \cos(k_y - q_y)\cos q_x. \tag{D.36}$$

Putting all parts together, we obtain the total scattering amplitude (4.77).

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