1 Scattering of light by an array of atoms

In our experiment we observe the scattering of photons from atoms confined in an optical lattice, here we treat this situation by obtaining the the field scattered from a single atom and then summing the field contributions from all the atoms coherently at the location of our detector. The main goal of this document is to find the connection between the intensity that we measure in our cameras and the spin structure factor as it is calculated by the theorists in our collaboration.

1.1 Electric field and intensity due to a single atom

To calculate the scattered field, one uses the source-field expression, which relates the radiated field to the emitting dipole moment, this is derived in the standard textbooks [1, 2]. The field at the position of the detector $\boldsymbol{r}_D$ is given by

$$E^{(+)}(\boldsymbol{r}_D, t) = \eta e^{-i\omega_L(t-r_D/c)} S_-\left(t - \frac{r_D}{c}\right) \tag{1}$$

where η is a proportionality factor that we will address later on. The time-averaged intensity at the detector is

$$\begin{aligned} \langle I(t)\rangle =& \langle E^{(-)}(\boldsymbol{r}_D, t) E^{(+)}(\boldsymbol{r}_D, t)\rangle \\ =& |\eta|^2 \langle S_+(t - r_D/c) S_-(t - r_D/c)\rangle \\ =& |\eta|^2 \langle S_+ S_-\rangle \\ =& |\eta|^2 \rho_{ee} \end{aligned} \tag{2}$$

In the third line the time dependence is dropped since we are interested in the steady state solution. We can write the $S_\pm$ operators of the atoms as

$$S_\pm = \langle S_\pm\rangle + \delta S_\pm \tag{3}$$

which defines the difference, δS, between $S_\pm$ and its average value. Writing $S_\pm$ this way allows us to distinguish between two components in the radiated light, the radiation of the average dipole $\langle S_\pm\rangle$ which is the radiation of a classical oscillating dipole with a phase that is well defined relative to the incident laser field, and the radiation form the $\delta S_\pm$ component which does not have a phase that is well defined relative to the incident field because it comes form the fluctuating part of the atomic dipole. The intensity is then a sum of coherent and incoherent parts

$$I = \eta^2 \langle S_+\rangle\langle S_-\rangle + \eta^2 \langle \delta S_+ \delta S_-\rangle \tag{4}$$

where we have used the fact that by definition $\langle \delta S_\pm\rangle = 0$. The first and second terms of this equation are the coherent and incoherent intensity which can be calculated by using the steady-state solutions to the optical Bloch equations given by

$$\langle S_\pm\rangle = u \pm iv \tag{5}$$

$$u = \frac{\Delta}{\Gamma\sqrt{I_\mathrm{p}/I_\mathrm{sat}}} \frac{s}{1+s} \tag{6}$$

$$v = \frac{1}{2\sqrt{I_\mathrm{p}/I_\mathrm{sat}}} \frac{s}{1+s} \tag{7}$$

$$\tag{8}$$

where s is the saturation parameter for an incident probe with intensity I_p:

$$s = \frac{2I_\mathrm{p}/I_\mathrm{sat}}{1+4(\Delta/\Gamma)^2} = \frac{s_0}{1+4(\Delta/\Gamma)^2} \tag{9}$$

and we have defined $s_0 = 2I_\mathrm{p}/I_\mathrm{sat}$. The coherent and incoherent intensities are

$$\begin{aligned} \frac{1}{\eta^2} I_\mathrm{coh} =& \frac{1}{2}\frac{s}{(1+s)^2} = \rho_{ee}\frac{1}{1+s} \\ \frac{1}{\eta^2} I_\mathrm{incoh} =& \langle S_+ S_- \rangle - \langle S_+\rangle\langle S_-\rangle = \frac{1}{2}\frac{s^2}{(1+s)^2} = \rho_{ee}\frac{s}{1+s} \end{aligned} \tag{10}$$

Note that if we add up coherent and incoherent part we get the more familiar result $I = \eta^2 \rho_{ee}$, where the total intensity is simply proportional to the population of the excited state.

1.2 Scattering cross-section

Now we will turn onto the evaluation of η, the proportionality factor between the field and the emitting dipole. Knowledge of η will allow us to sum coherently the field from a collection of atoms.

We start by considering the transition matrix element between the following initial and final states of the atom+photon system:

$$\begin{aligned} |\varphi_i\rangle =& |g; \boldsymbol{k}\boldsymbol{\varepsilon}\rangle \\ |\varphi_f\rangle =& |g; \boldsymbol{k}'\boldsymbol{\varepsilon}'\rangle \end{aligned} \tag{11}$$

These states represent the absorption and re-emission of a single photon by the atom, with two possibly different initial and final photon states.

The transition rate to from $i \to f$ is given by

$$w_{fi} = \frac{2\pi}{\hbar}|\mathcal{T}_{fi}|^2 \delta(E_f - E_i) \tag{12}$$

Where we use the notation in [2] (see Exercise 5 on pg. 530), and $\mathcal{T}_{fi}$ is given by

$$\mathcal{T}_{fi} = \frac{\langle g; \boldsymbol{k}'\boldsymbol{\varepsilon}'|H_I'|e;0\rangle\langle e;0|H_I'|g;\boldsymbol{k}\boldsymbol{\varepsilon}\rangle}{\hbar\omega - \hbar\omega_0 + i\hbar(\Gamma/2)} \tag{13}$$

where H_I' is the interaction Hamiltonian

$$H_I' = -\boldsymbol{d}\cdot\boldsymbol{E}_\perp(\boldsymbol{r}) \tag{14}$$

and[1]

$$\boldsymbol{E}_\perp(\boldsymbol{r}) = i\sum_j \left[\frac{\hbar\omega_j}{2\varepsilon_0 L^3}\right]^{1/2}\left(\hat{a}_j\boldsymbol{\varepsilon}_j e^{i\boldsymbol{k}_j\cdot\boldsymbol{r}} - \hat{a}_j^+\boldsymbol{\varepsilon}_j^* e^{-i\boldsymbol{k}_j\cdot\boldsymbol{r}}\right) \tag{15}$$

Notice that there is an intermediate excited state $|e;0\rangle$, since the absorption-emission event is a second order process.

Using the expressions for H_I' and $\boldsymbol{E}_\perp(\boldsymbol{r})$ we obtain for the matrix element

$$\langle e;0|H_I'|g;\boldsymbol{k}\boldsymbol{\varepsilon}\rangle = -i\sqrt{\frac{\hbar\omega}{2\varepsilon_0 L^3}}\langle e|(\boldsymbol{d}\cdot\boldsymbol{\varepsilon}^*)e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}|g\rangle \tag{16}$$

[1] Notice that the presence of ϵ_0 reveals that we are using SI units, following the treatment in [2].

At this point the textbook treatment usually assumes that the atom is at the origin and that the size of the atom wavefunction is very small compared to $|\boldsymbol{k}|^{-1}$, and so the exponential inside the matrix element typically does not show up. In our case the atom is in a lattice site and its center of mass state is one of the harmonic oscillator states of a lattice well is large enough that the exponential term cannot be neglected.

The states $|e\rangle$ and $|g\rangle$ include the center of mass and internal states of the atom. We separate the center of mass part, and keep the labels e, g for the internal states. Also, we denote the center of mass initial and final states as $|u\rangle$ and $|u'\rangle$ respectively, and the center of mass state of the intermediate excited state as $|v\rangle$, we have

$$\langle e;0|H_I'|g;\boldsymbol{k}\boldsymbol{\varepsilon}\rangle = -i\sqrt{\frac{\hbar\omega}{2\varepsilon_0 L^3}}\langle e|\boldsymbol{d}\cdot\boldsymbol{\varepsilon}^*|g\rangle\langle v|e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}|u\rangle \tag{17}$$

and similarly

$$\langle g;\boldsymbol{k}'\boldsymbol{\varepsilon}'|H_I'|e;0\rangle = i\sqrt{\frac{\hbar\omega'}{2\varepsilon_0 L^3}}\langle g|\boldsymbol{d}\cdot\boldsymbol{\varepsilon}'|e\rangle\langle u'|e^{i\boldsymbol{k}'\cdot\boldsymbol{r}}|v\rangle \tag{18}$$

This gives for the matrix element

$$\mathcal{T}_{fi} = \sum_v \frac{\sqrt{\omega\omega'}}{2\varepsilon_0 L^3}\frac{\langle g|\boldsymbol{d}\cdot\boldsymbol{\varepsilon}'|e\rangle\langle e|\boldsymbol{d}\cdot\boldsymbol{\varepsilon}^*|g\rangle\langle u'|e^{i\boldsymbol{k}'\cdot\boldsymbol{r}}|v\rangle\langle v|e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}|u\rangle}{\omega-\omega_0+i(\Gamma/2)} \tag{19}$$

where we have summed over all possible intermediate center of mass states. Note that the sum can be taken out using the closure relation $\sum_v |v\rangle\langle v| = \mathbb{1}$.

In our experiment we are driving a sigma-minus transition so we can consider only the projection of $\boldsymbol{d}$ onto $\boldsymbol{\varepsilon}_-$

$$\langle e|\boldsymbol{d}\cdot\boldsymbol{\varepsilon}^*|g\rangle \equiv d_-(\boldsymbol{\varepsilon}_-\cdot\boldsymbol{\varepsilon}^*) \tag{20}$$

which leads to

$$\mathcal{T}_{fi} = \frac{\sqrt{\omega\omega'}}{2\varepsilon_0 L^3}\frac{|d_-|^2(\boldsymbol{\varepsilon}_+\cdot\boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^*\cdot\boldsymbol{\varepsilon}_-)}{\omega-\omega_0+i(\Gamma/2)}\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle \tag{21}$$

We use the relation between $|d_-|^2$ and the linewidth of the transition

$$|d_-|^2 = 3\pi\varepsilon_0\hbar\left(\frac{c}{\omega_0}\right)^3\Gamma \tag{22}$$

and also the approximation $\omega' \approx \omega \approx \omega_0$ for the square root in the denominator to obtain

$$\mathcal{T}_{fi} = \frac{3}{k^2}\frac{\pi\hbar c}{L^3}(\boldsymbol{\varepsilon}_+\cdot\boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^*\cdot\boldsymbol{\varepsilon}_-)\frac{\Gamma/2}{\omega-\omega_0+i(\Gamma/2)}\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle \tag{23}$$

To obtain the scattering rate of photons towards a certain solid angle Ω', we must sum over all values of k' in the direction of Ω'. The number of final states with energy between $\hbar ck'$ and $\hbar c(k'+\mathrm{d}k')$ whose wave vector points inside the solid angle $\mathrm{d}\Omega'$ equals

$$\rho(\hbar ck')\hbar c\mathrm{d}k'\mathrm{d}\Omega' = \frac{L^3}{8\pi^3}k'^2\mathrm{d}k'\mathrm{d}\Omega' \tag{24}$$

where ρ is the density of states, which is a function of the photon energy $\hbar ck$. We use the density of states to replace the sum over k' with an integral, and obtain the total transition rate in the direction Ω':

$$\begin{aligned}\sum_f w_{fi} =& \frac{2\pi}{\hbar}\mathrm{d}\Omega'\int_0^\infty \frac{k'^2\mathrm{d}k'}{(2\pi/L^3)^3}|\mathcal{T}_{fi}|^2\delta(\hbar ck'-\hbar ck)\\ =&\mathrm{d}\Omega'\frac{9}{4k^2}\frac{c}{L^3}|(\boldsymbol{\varepsilon}_+\cdot\boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^*\cdot\boldsymbol{\varepsilon}_-)|^2\left|\frac{\Gamma/2}{\omega-\omega_0+i(\Gamma/2)}\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle\right|^2\\ =&\mathrm{d}\Omega'\frac{9}{4k^2}\frac{c}{L^3}|(\boldsymbol{\varepsilon}_+\cdot\boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^*\cdot\boldsymbol{\varepsilon}_-)|^2\frac{(\Gamma/2)^2}{\Delta^2+(\Gamma/2)^2}\left|\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle\right|^2\end{aligned} \tag{25}$$

If we consider the flux corresponding to the state of the initial photon $\phi = c/L^3$ then we can define the differential cross section

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega'} = \frac{\sum_f w_{fi}}{\mathrm{d}\Omega'\phi} = \frac{9}{4k^2}|(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^* \cdot \boldsymbol{\varepsilon}_-)|^2 \frac{(\Gamma/2)^2}{\Delta^2 + (\Gamma/2)^2} \left|\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle\right|^2 \tag{26}$$

From here we can write down the intensity on a detector located at $\boldsymbol{r}_D$ in the direction of $\mathrm{d}\Omega'$ as [2]

$$\begin{aligned} I =& \frac{1}{r_D^2}\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega'} I_\mathrm{p} = \frac{1}{r_D^2}\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega'}\frac{\hbar c k^3 \Gamma}{6\pi}\frac{I_\mathrm{p}}{I_\mathrm{sat}} \\ =& \frac{\hbar c k \Gamma}{r_D^2}\frac{9}{4(6\pi)}|(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^* \cdot \boldsymbol{\varepsilon}_-)|^2 \left|\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle\right|^2 \frac{s_0/2}{4(\Delta/\Gamma)^2+1} \end{aligned} \tag{27}$$

We identify the last fraction in this product as ρ_{ee} (in the limit of low intensity). Comparing with Eq. 10 we can write down an expression for η,

$$\eta = \left[\frac{\hbar c k \Gamma}{r_D^2}\frac{9}{24\pi}\right]^{1/2} (\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^* \cdot \boldsymbol{\varepsilon}_-)\langle u'|e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot\boldsymbol{r}}|u\rangle \tag{28}$$

With an exact expression for η we can obtain the field radiated by each atom and proceed to sum the field coherently for a collection of atoms.

1.3 Summation for a collection of atoms

For a collection of atoms, the resulting field is the sum of the field produced by each individual atom, so we have

$$\langle I(t)\rangle = \left\langle \left(\sum_m E_m^{(-)}(\boldsymbol{r}_D, t)\right)\left(\sum_n E_n^{(+)}(\boldsymbol{r}_D, t)\right)\right\rangle \tag{29}$$

where we have labeled the atoms with the indices m and n. We insert the source-field expression from Eq. 1 (dropping the time dependence)

$$I = \sum_{mn} \eta_m \eta_n^* \langle S_{m+} S_{n-}\rangle \tag{30}$$

Using $S = \langle S\rangle + \delta S$, as we did above to obtain the coherent and incoherent parts of the intensity, we obtain

$$\begin{aligned} I =& \sum_{mn} \eta_m \eta_n^* \left(\langle S_{m+}\rangle\langle S_{n-}\rangle + \langle \delta S_{m+}\delta S_{n-}\rangle\right) \\ =& \sum_{mn} \eta_m \eta_n^* \langle S_{m+}\rangle\langle S_{n-}\rangle + \sum_n |\eta_n|^2 \langle \delta S_{n+}\delta S_{n-}\rangle \end{aligned} \tag{31}$$

The steady state solutions of the optical Bloch equations are used again to evaluate the expectation values and we obtain for I

$$\begin{aligned} I = \sum_{mn} \eta_m \eta_n^* &\left(\frac{\Delta_m}{\Gamma\sqrt{I_\mathrm{p}/I_\mathrm{sat}}}\frac{s_m}{1+s_m} + i\frac{1}{2\sqrt{I_\mathrm{p}/I_\mathrm{sat}}}\frac{s_m}{1+s_m}\right)\left(\frac{\Delta_n}{\Gamma\sqrt{I_\mathrm{p}/I_\mathrm{sat}}}\frac{s_n}{1+s_n} - i\frac{1}{2\sqrt{I_\mathrm{p}/I_\mathrm{sat}}}\frac{s_n}{1+s_n}\right) \\ &+ \sum_n |\eta_n|^2 \frac{1}{2}\frac{s_n^2}{(1+s_n)^2} \end{aligned} \tag{32}$$

[2] Later on we will sum over output polarizations and final center of mass states of the atom, since our detection is insensitive to them.

$$I \;=\; \sum_{mn} \eta_m \eta_n^* \frac{s_m s_n}{(I_\mathrm{p}/I_\mathrm{sat})(1+s_m)(1+s_n)} \left(\frac{\Delta_m \Delta_n}{\Gamma^2} + i\frac{\Delta_n}{2\Gamma} - i\frac{\Delta_m}{2\Gamma} + \frac{1}{4} \right) \;+\; \sum_n |\eta_n|^2 \frac{1}{2}\frac{s_n^2}{(1+s_n)^2} \tag{33}$$

The last term here is the incoherently scattered part due to the fluctuating fraction $\delta S_\pm$ of the atomic dipole. The cross terms do not appear in this sum because $\langle \delta S_{m+} \delta S_{n-} \rangle = 0$ for $m \neq n$, this is in fact why this part is identified as the incoherent scattering.

We proceed to split up the first sum into same-atom ($n = m$) and different atom ($n < m$) parts

$$\begin{aligned} I = \sum_{m<n} \frac{s_m s_n}{(I_\mathrm{p}/I_\mathrm{sat})(1+s_m)(1+s_n)} &\left(\eta_m \eta_n^* \left(\frac{\Delta_m \Delta_n}{\Gamma^2} + i\frac{\Delta_n}{2\Gamma} - i\frac{\Delta_m}{2\Gamma} + \frac{1}{4} \right) \right. \\ &\left. + \eta_n \eta_m^* \left(\frac{\Delta_n \Delta_m}{\Gamma^2} + i\frac{\Delta_m}{2\Gamma} - i\frac{\Delta_n}{2\Gamma} + \frac{1}{4} \right) \right) \\ &+ \sum_n |\eta_n|^2 \frac{s_n s_n}{(I_\mathrm{p}/I_\mathrm{sat})(1+s_n)(1+s_n)} \left(\frac{\Delta_n \Delta_n}{\Gamma^2} + \frac{1}{4} \right) \\ &+ \sum_n |\eta_n|^2 \frac{1}{2}\frac{s_n^2}{(1+s_n)^2} \end{aligned} \tag{34}$$

$$\begin{aligned} I = \sum_{m<n} \frac{s_m s_n}{(I/I_\mathrm{sat})(1+s_m)(1+s_n)} 2\Re &\left[\eta_m \eta_n^* \left(\frac{\Delta_m \Delta_n}{\Gamma^2} + i\frac{\Delta_n}{2\Gamma} - i\frac{\Delta_m}{2\Gamma} + \frac{1}{4} \right) \right] \\ &+ \sum_n |\eta_n|^2 \frac{1}{2}\frac{s_n}{(1+s_n)^2} + \sum_n |\eta_n|^2 \frac{1}{2}\frac{s_n^2}{(1+s_n)^2} \end{aligned} \tag{35}$$

With this expression in hand we focus our attention on the terms $\eta_m \eta_n^*$ and $|\eta_n|^2$. We start with the latter

$$|\eta_n|^2 = \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} |(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^* \cdot \boldsymbol{\varepsilon}_-)|^2 \langle u | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u' \rangle \langle u' | e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u \rangle \tag{36}$$

and notice that we have to sum over output polarizations $\boldsymbol{\varepsilon}'$ and final center of mass states u', since our detector does not care about either. We obtain

$$\begin{aligned} \sum_{\boldsymbol{\varepsilon}' u'} |\eta_n|^2 =& \sum_{u'} \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \sum_{\boldsymbol{\varepsilon}'} |(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon}^* \cdot \boldsymbol{\varepsilon}_-)|^2 \langle u | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u' \rangle \langle u' | e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u \rangle \\ =& \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \langle u | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u \rangle \\ =& \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \end{aligned} \tag{37}$$

where we have used the closure relation $\sum u' | u' \rangle \langle u' | = \mathbb{1}$, and have defined for brevity

$$\Lambda = \sum_{\boldsymbol{\varepsilon}'} |(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}_-)|^2 \tag{38}$$

Similarly, for $\eta_m \eta_n^*$

$$\sum_{\boldsymbol{\varepsilon}' u_m' u_n'} \eta_m \eta_n^* = \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \sum_{u_m' u_n'} \langle u_n | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | u_n' \rangle \langle u_m' | e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_m} | u_m \rangle \tag{39}$$

In this case we cannot use the closure relation because n, m refer to different atoms. We simplify the treatment by considering only final states for the atom that are the same as the initial state $u' = u$ (these are going to have the largest matrix elements anyways). In the sum over u'_m, u'_n only $u'_m = u_m$ and $u'_n = u_n$ contribute. We take the center of mass state of the atoms to be the ground state of the single lattice site harmonic oscillator. This leaves us with

$$\sum_{\boldsymbol{\varepsilon}'} \eta_m \eta_n^* = \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \langle 0_n | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | 0_n \rangle \langle 0_m | e^{i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_m} | 0_m \rangle \tag{40}$$

1.3.1 Debye-Waller factor

For each center of mass expectation value we perform a translation $\boldsymbol{R}_n$ of the coordinate system such that the position of the n^{th} atom has a zero expectation value $\langle \boldsymbol{r}_n \rangle = 0$. A phase factor comes out that depends on the position $\boldsymbol{R}_n$ of the lattice site in which the atom is located:

$$\langle 0_n | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | 0_n \rangle = e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{R}_n} \langle 0_n | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | 0_n \rangle \tag{41}$$

We then use the equality $\langle e^{\hat{A}} \rangle = e^{\frac{1}{2}\langle \hat{A}^2 \rangle}$, which is valid for a simple harmonic oscillator, where $\hat{A}$ is any linear combination of displacement and momentum operators of the oscillator. This leaves us with

$$\begin{aligned}
\langle 0_n | e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n} | 0_n \rangle =& e^{-i(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{R}_n} e^{-\frac{1}{2}\left\langle [(\boldsymbol{k}'-\boldsymbol{k})\cdot \boldsymbol{r}_n]^2 \right\rangle} \\
=& e^{-i\boldsymbol{Q}\cdot \boldsymbol{R}_n} e^{-\frac{1}{2}\left\langle [\boldsymbol{Q}\cdot \boldsymbol{r}_n]^2 \right\rangle} \\
=& e^{-i\boldsymbol{Q}\cdot \boldsymbol{R}_n} \prod_{i=x,y,z} e^{-\frac{1}{2} Q_i^2 \langle r_{ni}^2 \rangle} \\
=& e^{-i\boldsymbol{Q}\cdot \boldsymbol{R}_n} e^{-W}
\end{aligned} \tag{42}$$

where we have defined the momentum transfer $\boldsymbol{Q} = \boldsymbol{k}' - \boldsymbol{k}$, and the Debye-Waller factor e^{-2W}.

Putting this back in the expression for $\eta_m \eta_n^*$ we get

$$\sum_{\boldsymbol{\varepsilon}'} \eta_m \eta_n^* = \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda e^{i\boldsymbol{Q}(\boldsymbol{R}_m - \boldsymbol{R}_n)} e^{-2W} \tag{43}$$

And if we now return to the expression for the intensity at the detector we have

$$\begin{aligned}
I = \sum_{m<n} \frac{s_m s_n}{(I_{\mathrm{p}}/I_{\mathrm{sat}})(1+s_m)(1+s_n)} 2\Re \left[\frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda e^{i\boldsymbol{Q}(\boldsymbol{R}_m - \boldsymbol{R}_n)} e^{-2W} \left(\frac{\Delta_m \Delta_n}{\Gamma^2} + i\frac{\Delta_n}{2\Gamma} - i\frac{\Delta_m}{2\Gamma} + \frac{1}{4} \right) \right] \\
+ \sum_n \frac{1}{2} \frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \frac{s_n}{1+s_n}
\end{aligned} \tag{44}$$

$$\begin{aligned}
I = &\left(\frac{\hbar c k \Gamma}{r_D^2} \frac{9}{24\pi} \Lambda \right) \times \\
&\sum_{m<n} \frac{s_m s_n}{(I_{\mathrm{p}}/I_{\mathrm{sat}})(1+s_m)(1+s_n)} 2\Re \left[e^{i\boldsymbol{Q}(\boldsymbol{R}_m - \boldsymbol{R}_n)} e^{-2W} \left(\frac{\Delta_m \Delta_n}{\Gamma^2} + i\frac{\Delta_n}{2\Gamma} - i\frac{\Delta_m}{2\Gamma} + \frac{1}{4} \right) \right] + \sum_n \frac{1}{2} \frac{s_n}{1+s_n}
\end{aligned} \tag{45}$$

It is good to see that for a large time-of-flight, where the Debye-Waller factor goes to zero due to large extent of the expanding atom wavefunctions, this formula reduces to the the standard uncorrelated scattering for N atoms, $I = N\rho_{ee}$ with $\rho_{ee} = \frac{1}{2}\frac{s}{1+s}$.

Note: To see this more clearly and at the same time check the prefactors that show up in this expression, we can evaluate the total photon scattering rate $\Gamma_{\rm scatt} = \frac{1}{\hbar ck}\int I r_D^2 \mathrm{d}\Omega$, for which we use $\int \Lambda \mathrm{d}\Omega = \frac{8\pi}{3}$ to obtain

$$\Gamma_{\rm scatt} = \Gamma \frac{1}{2}\frac{s}{1+s} = \Gamma \rho_{ee} \tag{46}$$

1.4 Large detuning limit

We start from Eq. (45) and concentrate on the two sums, the first of which is

$$\frac{e^{-2W}}{2I_{\rm p}/I_{\rm sat}} \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{s_m s_n}{(1+s_m)(1+s_n)} \left(4\Delta_m\Delta_n + 2i\Delta_n - 2i\Delta_m + 1\right) \tag{47}$$

where for simplicity we have now written the detunings in units of Γ. We will split this up further into four terms

$$\frac{e^{-2W}}{2I_{\rm p}/I_{\rm sat}} \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{s_m s_n}{(1+s_m)(1+s_n)} 4\Delta_m\Delta_n \tag{48}$$

$$\frac{e^{-2W}}{2I_{\rm p}/I_{\rm sat}} \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{s_m s_n}{(1+s_m)(1+s_n)} 2i\Delta_n \tag{49}$$

$$-\frac{e^{-2W}}{2I_{\rm p}/I_{\rm sat}} \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{s_m s_n}{(1+s_m)(1+s_n)} 2i\Delta_m \tag{50}$$

$$\frac{e^{-2W}}{2I_{\rm p}/I_{\rm sat}} \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{s_m s_n}{(1+s_m)(1+s_n)} \tag{51}$$

For a detuning such that $4\Delta_m^2, 4\Delta_n^2 \gg 1$ we have

$$\frac{s}{1+s} \approx \frac{2I_{\rm p}/I_{\rm sat}}{4\Delta^2 + 2I_{\rm p}/I_{\rm sat}} \tag{52}$$

and the four terms above go respectively to

$$e^{-2W} 2(I_{\rm p}/I_{\rm sat}) \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{4\Delta_m\Delta_n}{(4\Delta_m^2 + 2I_{\rm p}/I_{\rm sat})(4\Delta_n^2 + 2I_{\rm p}/I_{\rm sat})} \tag{53}$$

$$e^{-2W} 2(I_{\rm p}/I_{\rm sat}) \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{2i\Delta_n}{(4\Delta_m^2 + 2I_{\rm p}/I_{\rm sat})(4\Delta_n^2 + 2I_{\rm p}/I_{\rm sat})} \tag{54}$$

$$-e^{-2W} 2(I_{\rm p}/I_{\rm sat}) \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{2i\Delta_m}{(4\Delta_m^2 + 2I_{\rm p}/I_{\rm sat})(4\Delta_n^2 + 2I_{\rm p}/I_{\rm sat})} \tag{55}$$

$$e^{-2W} 2(I_{\rm p}/I_{\rm sat}) \Re \sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \frac{1}{(4\Delta_m^2 + 2I_{\rm p}/I_{\rm sat})(4\Delta_n^2 + 2I_{\rm p}/I_{\rm sat})} \tag{56}$$

Furthermore, if we detune the light in between the two spin states then we can use, $\Delta_m^2 = \Delta^2$ and

$\Delta_m = 2|\Delta|S_{zm}$, where $S_{zm} = \pm\frac{1}{2}$ is the spin state of the atom in site m, to obtain

$$e^{-2W}2(I_\mathrm{p}/I_\mathrm{sat})\frac{16\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})^2}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} \tag{57}$$

$$e^{-2W}2(I_\mathrm{p}/I_\mathrm{sat})\frac{4i|\Delta|}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})^2}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zn} \tag{58}$$

$$-e^{-2W}2(I_\mathrm{p}/I_\mathrm{sat})\frac{4i|\Delta|}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})^2}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm} \tag{59}$$

$$e^{-2W}2(I_\mathrm{p}/I_\mathrm{sat})\frac{1}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})^2}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \tag{60}$$

We identify the first term and the last term as related to the spin structure factor and crystal structure factor. In this last equation we see that the first term, the one related to the spin structure factor, is going to have the main contribution to the intensity because it goes as $|\Delta|^{-2}$, whereas the other terms go as larger powers of $1/|\Delta|$. If we neglect terms other than the first one, we obtain

$$\begin{aligned} I &= \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\left[e^{-2W}\frac{16\Delta^2(I_\mathrm{p}/I_\mathrm{sat})}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})^2}2\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn}+\sum_n\frac{1}{2}\frac{2I_\mathrm{p}/I_\mathrm{sat}}{4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat}}\right] \\ &= \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\frac{I_\mathrm{p}/I_\mathrm{sat}}{4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat}}\left[\frac{e^{-2W}16\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})}2\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn}+N\right] \end{aligned} \tag{61}$$

We note that, since exchanging indexes in the summand results in its complex conjugate, $2\Re\sum_{m<n}\equiv\sum_{m\neq n}$. We also use $\sum_{m\neq n}\equiv\sum_{mn}-\sum_{m=n}$ to obtain

$$\begin{aligned} I &= \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\frac{I_\mathrm{p}/I_\mathrm{sat}}{4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat}}\left[\frac{e^{-2W}16\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})}\left(\sum_{mn}-\sum_{m=n}\right)e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn}+N\right] \\ &= \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\frac{I_\mathrm{p}/I_\mathrm{sat}}{4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat}}\left[\frac{e^{-2W}4\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})}\left(4\sum_{mn}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn}-N\right)+N\right] \end{aligned} \tag{62}$$

At this point we consider a measurement of the intensity after a large time-of-flight (TOF), denoted as I_∞. After TOF, the Debye-Waller factor goes to zero due to the expanding size of the atomic wavefunction, so we have

$$I_\infty = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\frac{I_\mathrm{p}/I_\mathrm{sat}}{4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat}}N \tag{63}$$

$$\frac{I}{I_\infty} = \frac{e^{-2W}4\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})}\left(\frac{4}{N}\sum_{mn}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn}-1\right)+1 \tag{64}$$

1.5 Measurement of the structure factor

The theorists in our collaboration calculate the spin structure factor $S(\boldsymbol{Q})$, which is defined as

$$S(\boldsymbol{Q}) = \frac{4}{N}\sum_{mn}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} \tag{65}$$

From inspection of Eq. 64 we see that a measurement of I/I_∞ can be related to the spin structure factor by

$$\frac{I}{I_\infty} = \frac{e^{-2W}4\Delta^2}{(4\Delta^2+2I_\mathrm{p}/I_\mathrm{sat})}\left(S(\boldsymbol{Q})-1\right)+1 \tag{66}$$

At this point we introduce the notation

$$\frac{I}{I_\infty} \equiv I_{\boldsymbol{Q}} \qquad\qquad c \equiv \frac{e^{-2W}4\Delta^2}{(4\Delta^2 + 2I_{\mathrm{p}}/I_{\mathrm{sat}})} \qquad\qquad S(\boldsymbol{Q}) \equiv S_{\boldsymbol{Q}} \tag{67}$$

$$I_{\boldsymbol{Q}} = c(S_{\boldsymbol{Q}} - 1) + 1 \tag{68}$$

If we perform the measurement in a deep enough lattice, then the Debye-Waller factor $e^{-2W} \to 1$. Furthermore, if we select an intensity such that $I_{\mathrm{p}}/I_{\mathrm{sat}} \ll 2\Delta^2$ this reduces to an equivalence between the TOF normalized intensity and the spin structure factor:

$$I_{\boldsymbol{Q}} = S_{\boldsymbol{Q}} \tag{69}$$

To compare with the calculations from the theorists we solve for $S_{\boldsymbol{Q}}$ and find

$$S_{\boldsymbol{Q}} = 1 + (I_{\boldsymbol{Q}} - 1)/c \tag{70}$$

In our experiment we are using $I_{\mathrm{p}}/I_{\mathrm{sat}} \approx 15$ and $\Delta = 6.5$. We perform the measurement at a lattice depth of $50E_R$. For this parameters we have the following:

$$\begin{aligned} e^{-2W} &= 0.81 \\ 1 + \frac{I_{\mathrm{p}}/I_{\mathrm{sat}}}{2\Delta^2} &= 1.18 \end{aligned} \tag{71}$$

$$S_{\boldsymbol{Q}} - 1 = 1.45(I_t - 1) \tag{72}$$

1.6 Magneto-association and contributions from doubly occupied sites

In our experiment we have the possibility to magneto-associate (MA) the atoms in doubly occupied sites prior to a measurement of the scattered light. The formed molecules are insensitive to the probe light and do not scatter any light at all. In this section we study the implications of MA for the comparison between experimental measurements and the structure factor as calculated by the theorists.

We start from Eq. 62 and will separate the sums over atoms in singly and doubly occupied sites. We have for the intensity

$$\begin{aligned} I &= \frac{I_\infty}{N}\left[4c\left(\sum_{mn} - \sum_{m=n}\right) e^{i\boldsymbol{Q}(\boldsymbol{R}_m - \boldsymbol{R}_n)} S_{zm}S_{zn} + \sum_n\right] \\ &= \frac{I_\infty}{N}\left[4c\sum_{mn} e^{i\boldsymbol{Q}(\boldsymbol{R}_m - \boldsymbol{R}_n)} S_{zm}S_{zn} - c\sum_n + \sum_n\right] \end{aligned} \tag{73}$$

With MA prior to the intensity measurement, there is no contribution to the scattered light from atoms in doubly occupied sites, so denoting as $\mathcal{S}$ the set of singly occupied sites, we have

$$\begin{aligned} I_{\mathrm{MA}} &= \frac{I_\infty}{N}\left[4c\sum_{m\in\mathcal{S}} e^{i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm} \sum_{n\in\mathcal{S}} e^{-i\boldsymbol{Q}\boldsymbol{R}_n} S_{zn} - c\sum_{n\in\mathcal{S}} + \sum_{n\in\mathcal{S}}\right] \\ &= \frac{I_\infty}{N}\left[4c\sum_{m\in\mathcal{S}} e^{i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm} \sum_{n\in\mathcal{S}} e^{-i\boldsymbol{Q}\boldsymbol{R}_n} S_{zn} - cN(1-D) + N(1-D)\right] \end{aligned} \tag{74}$$

where D is the fraction of atoms in doubly occupied sites.

Denoting the set of atoms in doubly occupied sites as $\mathcal{D}$, we point out that

$$\sum_{m\in\mathcal{D}} e^{\pm i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm} = 0 \tag{75}$$

since the contributions cancel out in pairs as $e^{\pm i\boldsymbol{Q}\boldsymbol{R}_m}(+1/2-1/2)=0$. This allows us to remove the $\in \mathcal{S}$ constraint in the remaining sums,

$$\sum_{m\in\mathcal{S}} e^{\pm i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm} = \sum_{m} e^{\pm i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm} \tag{76}$$

and obtain for the intensity after MA

$$\begin{aligned} I_{\mathrm{MA}} &= \frac{I_\infty}{N}\left[4c\sum_m e^{i\boldsymbol{Q}\boldsymbol{R}_m} S_{zm}\sum_n e^{-i\boldsymbol{Q}\boldsymbol{R}_n} S_{zn} - cN(1-D) + N(1-D)\right] \\ &= I_\infty\left[cS_{\boldsymbol{Q}} - c(1-D) + (1-D)\right] \end{aligned} \tag{77}$$

We introduce the simplifying notation

$$\frac{I_{\mathrm{MA}}}{I_\infty} \equiv I_{\boldsymbol{Q}m} \tag{78}$$

and simplify for the spin structure factor we obtain

$$S_{\boldsymbol{Q}} = \frac{1}{c}\left(I_{\boldsymbol{Q}m} - 1\right) - \frac{D}{c}(c-1) + 1 \tag{79}$$

To measure the fraction of atoms in doubly occupied sites, D, we can make a measurement of the scattered intensity after MA and a large TOF. Combined with the TOF measurement without MA we have

$$D = 1 - \frac{I_{\mathrm{MA},\infty}}{I_\infty} \equiv 1 - I_s \tag{80}$$

giving finally

$$S_{\boldsymbol{Q}} = \frac{1}{c}\left(I_{\boldsymbol{Q}m} - 1\right) + \frac{c-1}{c}\left(I_s - 1\right) + 1 \tag{81}$$

where we recall

$$I_{\boldsymbol{Q}m} = \frac{I_{\mathrm{MA}}}{I_\infty} \qquad\qquad I_s = \frac{I_{\mathrm{MA},\infty}}{I_\infty} \tag{82}$$

1.7 Data at short time-of-flight

In the above section we considered I_∞, a measurement of the intensity with a time-of-flight long enough such that the Debye-Waller factor $e^{-2W} \to 0$. For some of the data we have taken, the TOF is 6 μs, which in some cases is not long enough for the Debye-Waller factor be negligible. In this section we look at the determination of $S_{\boldsymbol{Q}}$ with time-of-flight data for which one cannot assume $e^{-2W} \to 0$.

We recall that the Debye-Waller factor is defined as

$$e^{-2W} = \prod_{i=x,y,z} e^{-Q_i^2\langle r_i^2\rangle} \tag{83}$$

where Q_i is the i^{th} component of the momentum transfer $\boldsymbol{Q} = \boldsymbol{k}' - \boldsymbol{k}$, with $\boldsymbol{k}'$ and $\boldsymbol{k}$ the wave vectors of the outgoing and incoming light respectively. The time-of-flight dependence of the Debye-Waller factor is through the expanding size of the atomic wavefunctions. For an in-situ picture $\langle r_i^2\rangle$ corresponds to the position spread of the lattice site's harmonic oscillator ground state. After the atoms are released from the lattice the spread of the wavefunction satisfies

$$\begin{aligned}\langle r_i^2\rangle_t =&\langle r_i^2\rangle_0 + \frac{t^2}{m^2}\langle p_i^2\rangle_0 \\ =&\langle r_i^2\rangle_0 + \frac{t^2}{m^2}\frac{\hbar^2}{4\langle r_i^2\rangle_0}\end{aligned} \tag{84}$$

In a harmonic oscillator potential we have

$$\langle r^2\rangle = \frac{\hbar}{2m\omega} \tag{85}$$

and since in a lattice of depth V_0 recoils, $\hbar\omega = 2E_R\sqrt{V_0}$

$$\langle r^2\rangle = \frac{a^2}{2\pi^2\sqrt{V_0}} \tag{86}$$

where a is the lattice spacing.

We then have for the Debye-Waller factor as a function of time-of-flight

$$\begin{aligned}[e^{-2W}]_t =& \prod_{i=x,y,z}\exp\left[-Q_i^2\left(\frac{a^2}{2\pi^2\sqrt{V_0}} + \frac{h^2\sqrt{V_0}}{8a^2}\frac{t^2}{m^2}\right)\right] \\ =&[e^{-2W}]_0\prod_{i=x,y,z}\exp\left[-Q_i^2\frac{h^2\sqrt{V_0}}{8a^2}\frac{t^2}{m^2}\right] \\ =&[e^{-2W}]_0\prod_{i=x,y,z}\exp\left[-\frac{\sqrt{V_0}}{2}\left(\frac{Q_i h}{2ma}\right)^2 t^2\right] \\ =&[e^{-2W}]_0\exp\left[-\frac{\sqrt{V_0}}{2}\left(\frac{|\boldsymbol{Q}|h}{2ma}\right)^2 t^2\right]\end{aligned} \tag{87}$$

We now define the time dependent correction factor

$$c(t) = \frac{[e^{-2W}]_t 4\Delta^2}{4\Delta^2 + 2I_\mathrm{p}/I_\mathrm{sat}} \tag{88}$$

The measured intensity after time-of-flight t is

$$\frac{I(t)}{I_\infty} = c(t)(S_{\boldsymbol{Q}} - 1) + 1 \tag{89}$$

Consider two measurements, one at $t = 0$, the other one at $t = T$,

$$\frac{I(t=0)}{I_\infty} = c(0)(S_{\boldsymbol{Q}} - 1) + 1 \qquad \frac{I(t=T)}{I_\infty} = c(T)(S_{\boldsymbol{Q}} - 1) + 1 \tag{90}$$

$$\frac{I(t=0)}{I(t=T)} = \frac{c(0)(S_{\boldsymbol{Q}} - 1) + 1}{c(T)(S_{\boldsymbol{Q}} - 1) + 1} \equiv I_{\boldsymbol{Q}}(T) \tag{91}$$

which can be solved to give

$$S_{\boldsymbol{Q}} = \frac{1 - c(0) - I_{\boldsymbol{Q}}(T)(1 - c(T))}{I_{\boldsymbol{Q}}(T)c(T) - c(0)} \tag{92}$$

1.8 Simultaneous measurement of the structure factor for two different values of $\boldsymbol{Q}$

In the sections above we have shown how to measure the spin structure factor $S_{\boldsymbol{Q}}$ by measuring the scattered intensity in-situ and after time-of-flight. An issue that arises with this measurement is that the in-situ and TOF measurements require two different realizations of the experiment.

In our setup we have the possibility of measuring the scattered intensity at two different output angles. One of the angles corresponds to a momentum transfer $\boldsymbol{Q} = \frac{2\pi}{a}(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2}) \equiv \boldsymbol{\pi}$, which offers the possibility of measuring the staggered magnetization of the system, i.e. the degree of antiferromagnetic ordering. The other angle corresponds to a momentum transfer $\boldsymbol{Q} = \frac{2\pi}{a}(0.4,\,-0.1,\,-0.04) \equiv \boldsymbol{\theta}$. The structure factor can also be measured at this value of $\boldsymbol{Q}$ and compared with theoretical calculations. Also, this momentum transfer does not measure the staggered magnetization, so it may be used as a normalization for the measurement at $\boldsymbol{Q} = \boldsymbol{\pi}$

For the measurements at $\boldsymbol{\pi}$ and $\boldsymbol{\theta}$ we have (without magneto-association)

$$S_{\boldsymbol{\pi}} = (I_{\boldsymbol{\pi}} - 1 + c)/c \qquad\qquad S_{\boldsymbol{\theta}} = (I_{\boldsymbol{\theta}} - 1 + c)/c \tag{93}$$

$$\frac{S_{\boldsymbol{\pi}}}{S_{\boldsymbol{\theta}}} = \frac{I_{\boldsymbol{\pi}} - 1 + c}{I_{\boldsymbol{\theta}} - 1 + c} \tag{94}$$

We recall that $I_{\boldsymbol{Q}} = I(\boldsymbol{Q})/I_\infty(\boldsymbol{Q})$. To make use of the simultaneous measurement of I at $\boldsymbol{Q} = \boldsymbol{\pi}, \boldsymbol{\theta}$ we define

$$\left(\frac{I(\boldsymbol{\pi})}{I(\boldsymbol{\theta})}\right) \Big/ \left(\frac{I_\infty(\boldsymbol{\pi})}{I_\infty(\boldsymbol{\theta})}\right) \equiv I_{\boldsymbol{\pi}/\boldsymbol{\theta}} \tag{95}$$

This measurement consists of an in-situ measurement of the ratio $I(\boldsymbol{\pi})/I(\boldsymbol{\theta})$ and a TOF measurement of the same ratio in a subsequent realization of the experiment. We then have

$$S_{\boldsymbol{\pi}} = S_{\boldsymbol{\theta}} \frac{I_{\boldsymbol{\pi}/\boldsymbol{\theta}} - (1-c)/I_{\boldsymbol{\theta}}}{1 - (1-c)/I_{\boldsymbol{\theta}}} \tag{96}$$

This measurement is an alternative to the measurement of $S_{\boldsymbol{\pi}}$ directly from $I_{\boldsymbol{\pi}}$, it offers the added advantage that the recording of the ratio $I(\boldsymbol{\pi})/I(\boldsymbol{\theta})$ in a single shot may result in a reduction of the noise in the signal.

From what we have seen so far, the noise in the signal is dominated by the repeatability of the experimental realizations. That is, the noise exceeds the detection noise (variance of the intensity measurement without atoms), and it also exceeds the shot to shot variation from atom number and probe intensity/polarization fluctuations. With this realization-dominated noise, a significant reduction in the error associated with $S_{\boldsymbol{\pi}}$ is not expected with the simultaneous measurement. Nevertheless, this alternate measure should be consistent with $S_{\boldsymbol{\pi}}$ as obtained directly from $I_{\boldsymbol{\pi}}$.

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