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

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

where 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:

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

The coherent and incoherent intensities are

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

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

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

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]

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

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

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

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, which, as we will see, 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. Here we separate the two, and keep the labels e, g for the internal states. Also, we denote the center of mass initial and final

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

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

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

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. We use this to replace the sum over k' with an integral, and obtain the total transition rate

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

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 (of light with polarization $\boldsymbol{\varepsilon}'$) on a detector located at $\boldsymbol{r}_D$ in the direction of $\mathrm{d}\Omega'$ as

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

We identify the last term in the product as ρ_{ee} (in the limit of low intensity). Comparing with Eq. 10 we can write down an expression for η which was defined back in Eq. (2),

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

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. **??** (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

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

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

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 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 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 $\Gamma_{\mathrm{scatt}} = \frac{1}{\hbar c k} \int I r_D^2 \mathrm{d}\Omega$, for which we use $\int \Lambda \mathrm{d}\Omega = \frac{8\pi}{3}$ to obtain

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

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

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

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

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

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

In the low intensity limit, and for a detuning such that $4\Delta_m^2, 4\Delta_n^2 \gg 1$ these four tend respectively to

$$e^{-2W}2(I_{\rm p}/I_{\rm sat})\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}\frac{1}{4\Delta_m\Delta_n} \tag{52}$$

$$e^{-2W}2(I_{\rm p}/I_{\rm sat})\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}\frac{i}{8\Delta_n\Delta_m^2} \tag{53}$$

$$-e^{-2W}2(I_{\rm p}/I_{\rm sat})\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}\frac{i}{8\Delta_n^2\Delta_m} \tag{54}$$

$$e^{-2W}2(I_{\rm p}/I_{\rm sat})\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}\frac{1}{16\Delta_m^2\Delta_n^2} \tag{55}$$

Furthermore, if we detune the light in between the two spin states then we can use $\frac{1}{2\Delta_m}=\frac{1}{|\Delta|}S_{zm}$, where $S_{zm}=\pm\frac{1}{2}$ is the spin state of the atom in site m, to obtain

$$e^{-2W}\frac{2I_{\rm p}/I_{\rm sat}}{|\Delta|^2}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} \tag{56}$$

$$e^{-2W}\frac{2I_{\rm p}/I_{\rm sat}}{4|\Delta|^3}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}iS_{zn} \tag{57}$$

$$-e^{-2W}\frac{2I_{\rm p}/I_{\rm sat}}{4|\Delta|^3}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}iS_{zm} \tag{58}$$

$$e^{-2W}\frac{2I/I_{\rm sat}}{16|\Delta|^4}\Re\sum_{m<n}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)} \tag{59}$$

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 spin structure factor then, in the low intensity and large detuning limit (with the detuning set in between the two spin states), we obtain

$$I = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right) \times \left(e^{-2W}\frac{2I_\mathrm{p}/I_\mathrm{sat}}{|\Delta|^2}\Re\sum_{m<n} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} + \frac{I_\mathrm{p}/I_{sat}}{4\Delta^2}N\right) \tag{60}$$

where we have made use of $s_n/(1+s_n) \approx (I_\mathrm{p}/I_\mathrm{sat})/\Delta_n^2$ to carry out the sum over n in Eq. (45). We then manipulate the $n < m$ sum and the real part to obtain

$$I = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right) \times \left(e^{-2W}\frac{I_\mathrm{p}/I_\mathrm{sat}}{|\Delta|^2}\sum_{mn} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} + (1-e^{-2W})\frac{I_\mathrm{p}/I_\mathrm{sat}}{4|\Delta|^2}N\right) \tag{61}$$

In this formula the spin structure factor appears explicitly, we pull out some factors and get

$$I = \frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\frac{I_\mathrm{p}/I_\mathrm{sat}}{4|\Delta|^2}N\left(1+e^{-2W}\left(\frac{4}{N}\sum_{mn} e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}S_{zm}S_{zn} - 1\right)\right) \tag{62}$$

After time-of-flight the Debye-Waller factor goes to zero due to the expanding size of the atomic wave-functions and so

$$I_\mathrm{TOF} = \frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\frac{I_\mathrm{p}/I_\mathrm{sat}}{4|\Delta|^2}N \tag{63}$$

giving finally

$$\frac{I}{I_\mathrm{TOF}} = 1 + e^{-2W}(S(\boldsymbol{Q})-1) \tag{64}$$

This has the expected form (we got this from David Huse), and it defines the spin structure factor as

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

IMPORTANT REMARK: We note here that this derivation which relates the observed intensity to the spin structure factor relies on the saturation parameter being much less than 1. In our case we have $I_\mathrm{p}/I_\mathrm{sat} \approx 25$ and $\Delta \approx 6.5$ which gives a saturation parameter of $s = 0.3$ which is less than 1, but maybe not entirely negligible. We have performed numerical evaluations of the exact expression for the intensity at the detector which considers saturation effects to determine what kind of corrections do we need to make to connect between our measurement and the exact spin structure factor (more on this below).

2 Numerical evaluation of the scattered intensity

For the numerical calculation we start from Eq. (45) and replace the sum over $m < n$ with an unrestricted sum over m, n. This removes the real part, and we have to subtract again some $m = n$ terms, the result is

$$I = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right) \times$$
$$\frac{e^{-2W}}{4(I_\mathrm{p}/I_\mathrm{sat})}\sum_{mn}\frac{s_m s_n}{(1+s_m)(1+s_n)}e^{i\boldsymbol{Q}(\boldsymbol{R}_m-\boldsymbol{R}_n)}\left(4\Delta_m\Delta_n + 2i\Delta_n - 2i\Delta_m + 1\right) + \sum_n \frac{1}{2}\frac{s_n}{1+s_n}\left(1-\frac{e^{-2W}}{1+s_n}\right) \tag{66}$$

We will split this up as

$$I = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right) \times$$
$$\left[\frac{e^{-2W}}{4(I_{\rm p}/I_{\rm sat})}\left(2\sum_m \frac{s_m}{1+s_m}\Delta_m e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} 2\sum_n \frac{s_n}{1+s_n}\Delta_n e^{-i\boldsymbol{Q}\cdot\boldsymbol{R}_n} \right.\right.$$
$$+ 2i\sum_m \frac{s_m}{1+s_m} e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} \sum_n \frac{s_n}{1+s_n}\Delta_n e^{-i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$$
$$- 2i\sum_m \frac{s_m}{1+s_m}\Delta_m e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} \sum_n \frac{s_n}{1+s_n} e^{-i\boldsymbol{Q}\cdot\boldsymbol{R}_n}$$
$$\left. + \sum_m \frac{s_m}{1+s_m} e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} \sum_n \frac{s_n}{1+s_n} e^{-i\boldsymbol{Q}\cdot\boldsymbol{R}_n}\right)$$
$$\left. + \sum_n \frac{1}{2}\frac{s_n}{1+s_n}\left(1 - \frac{e^{-2W}}{1+s_n}\right)\right] \tag{67}$$

The following sums appear and we define some shorthand notation for them

$$\Phi \equiv \sum_m \frac{s_m}{1+s_m}\Delta_m e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} \tag{68}$$

$$\Upsilon \equiv \sum_m \frac{s_m}{1+s_m} e^{i\boldsymbol{Q}\cdot\boldsymbol{R}_m} \tag{69}$$

$$\kappa \equiv \sum_n \frac{s_n}{1+s_n} \tag{70}$$

$$\xi \equiv \sum_n \frac{s_n}{(1+s_n)^2} \tag{71}$$

$$\tag{72}$$

We can then write the intensity as

$$I = \left(\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{24\pi}\Lambda\right)\left[\frac{e^{-2W}}{4(I_{\rm p}/I_{\rm sat})}\left(4\Phi\Phi^* + 2i\Upsilon\Phi^* - 2i\Phi\Upsilon^* + \Upsilon\Upsilon^*\right) + \frac{1}{2}\kappa - \frac{e^{-2W}}{2}\xi\right] \tag{73}$$

For the numerical evaluation we will use $\frac{\hbar ck\Gamma}{r_D^2}\frac{9}{48\pi}$ as a unit for the intensity, so we can finally simplify the expression to

$$I = \Lambda\left[\kappa + e^{-2W}\left(\frac{|\Upsilon - 2i\Phi|^2}{2(I_{\rm p}/I_{\rm sat})} - \xi\right)\right] \tag{74}$$

We recall that

$$\Lambda = \sum_{\boldsymbol{\varepsilon}'} |(\boldsymbol{\varepsilon}_+ \cdot \boldsymbol{\varepsilon}')(\boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}_-)|^2 \tag{75}$$

2.1 Evaluation in a lattice with AFM core and random shell

For the numerical evaluation of the scattered intensity we will consider a lattice with $L \times L \times L$ sites in which there is a core of size $L_{\rm AFM} \times L_{\rm AFM} \times L_{\rm AFM}$ in which the atoms have antiferromagnetically ordered spins. The distribution of the spins outside the core is random, but the spin imbalance is constrained to be zero, so there is an equal number of atoms in state $|1\rangle$ and state $|2\rangle$ occupying the L^3 sites in the lattice.

figures/bragg_setup_illustration_320px.png

Figure 1: Figure shows the different input directions that allow us to measure the spin structure factor at various values of the momentum transfer $\boldsymbol{Q}$.

2.1.1 Available values of momentum transfer $\boldsymbol{Q}$

In our experiment we sample four different values of the momentum transfer vector $\boldsymbol{Q}$ by sending the probe beam in from different viewports in our vacuum chamber. A schematic of this is shown in Fig. 1.

2.1.2 Debye-Waller factor

We will start by showing plots of the Debye-Waller factor as a function of time-of-flight and lattice depth for some of the momentum transfer values. Plots are as a function of lattice depth (Fig. 2) and also time-of-flight for a 20 recoil lattice (Fig. 3).

2.1.3 Saturation due to probe intensity

We look at the saturation due to the probe intensity for a momentum transfer $\boldsymbol{Q}_{\mathrm{AF}}$ corresponding to the HHH Bragg scattering peak for antiferromagnetically ordered sample, see Fig. 4. In this case we find that for the power that we are using, $\approx 250\,\mu$W, the excess of counts at the Bragg camera $A2$, for an in-situ picture with respect to a TOF picture, are underestimated by $\approx$11%.

../debye-waller_HHH/DWv0_plot.png

Figure 2: Figure shows the Debye-Waller factor as a function of lattice depth for different momentum transfers.

../debye-waller_HHH/DWtof_plot_20Er.png

Figure 3: Figure shows the Debye-Waller factor as a function of TOF for different momentum transfers. At $t = 0$ the atoms are released from a 20 recoil lattice.

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../pbragg_HHH/pbragg_HHH.png

Figure 4: We use the abbreviation $\frac{X}{X_{\mathrm{TOF}}} = X_t$. Left panel shows the Bragg scattering intensity at camera $A2$ for a beam on the HHH input (Input #3) as a function of probe power (Power translates into intensity given the known beam waist at the atoms). This corresponds to a momentum transfer equal to $\boldsymbol{Q}_{\mathrm{AF}}$. The various sets correspond to different sizes of the AFM core used in the numerical evaluation. Right panel shows the same as the left panel, but normalized to the value at zero power for each set. All the sets collapse into one, and the dependency due to power at low powers is fitted to a straight line.

At the moment we will carry on with the assumption that our measurement is in the low intensity regime, in this case we can go by Eq. (64) to get the value of the spin structure factor since we know the value of the Debye-Waller factors for each of the momentum transfers that we measure. Due to saturation effects then our value can be thought of as a lower bound to $S(\boldsymbol{Q})$. In the future we are going to try to repeat the measurements with a lower Bragg power so that we do not run into this kind of issues.

2.2 Estimating the size of the AFM domain

In our numerical evaluation of the scattered intensity we consider a lattice with an AFM domain in the center surrounded by a core with a random spin distribution. This allows us to study the effect of the outside shell on our ability to see an AFM distribution in the core, see Fig.5. Our system size is approximately $L = 40$, and for this size the Bragg signals that we obtain $A2_{\mathrm{t}} \approx 1.25$ (we have seen up to 1.5 sometimes) would be consistent with an AFM core size of approximately 6^3 sites. On the other hand we also see a depletion on camera $A1$ of $A1_t \approx 0.75$, which would be consistent with an AFM core of 25^3 lattice sites. We believe that our model is not really good for this kind of estimates and this is more the realm of the QMC people, in any case we show what we obtain.

2.3 Data with Debye-Waller factor correction for direct comparison with QMC

As was mentioned above, our QMC collaborators compute $S(\boldsymbol{Q})$, which is related to our measurement as

$$I_t = 1 + e^{-2W}(S(\boldsymbol{Q}) - 1) \tag{76}$$

where we use the abbreviation $I_t = \frac{I}{I_{\mathrm{TOF}}}$. Previously we presented our collaborators the data which did not have a Debye-Waller factor correction. Here we make a correction, also noting that we only made measurements in-situ, $T = 0$, and at $T = 6$ μs TOF. With these two measurements one can solve for

../crystalsize/CrystalSize.png

Figure 5: Left panel shows the intensity at camera $A1$ normalized to TOF for different system sizes and AFM core sizes. Right panel shows the same for the $A2$ camera.

the spin structure factor

$$\begin{aligned}
A &= \text{constant} \\
I_0 &= A(1 + DW_0(S(\boldsymbol{Q}) - 1)) \\
I_T &= A(1 + DW_T(S(\boldsymbol{Q}) - 1)) \\
I_t \equiv \frac{I_0}{I_T} &= \frac{1 + DW_0(S(\boldsymbol{Q}) - 1)}{1 + DW_T(S(\boldsymbol{Q}) - 1)} \\
\Rightarrow S(\boldsymbol{Q}) &= \frac{I_t(1 - DW_T) - (1 - DW_0)}{DW_0 - I_t DW_T}
\end{aligned} \tag{77}$$

The data including this Debye-Waller factor correction is shown in Fig. 6.

References

[1] R. Loudon, *The Quantum Theory of Light*, *Oxford Science Publications* (OUP Oxford, 2000).

[2] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications*, *A Wiley-Interscience publication* (Wiley, 1998).

figures/rocking.png

Figure 6: Top row shows our raw data, which corresponds to ratio of CCD counts $A2/A1$, and CCD counts for $A1$, $A2$ respectively. Middle row shows the ratio of in-situ and TOF data for each of the three quantities. Bottom row, left panel shows the Debye-Waller factor for the in-situ picture and the 6 μs TOF picture as a function of angle along chamber. The $A1$ Debye-Waller factor goes to 1 for both in-situ and TOF at 30°, which corresponds to zero momentum transfer. Bottom row, right panels show the spin structure factor determined from our data and the Debye-Waller factor corrections.