6 Wavepacket Coherence Transfer via DD interactions

We have shown that electron correlations, induced by controlled DD interactions, can enable the coherent transfer of electronic wavepacket motion from atoms to their neighbors. In the experiment, a 5 ns tunable dye laser excites Rb atoms in a MOT to the 25s state in a weak static electric field for which the tunable $25s33s \leftrightarrow 24p34p$ DD interaction is resonant. A picosecond THz pulse then further excites each Rydberg atom into a coherent superposition, of 25s and 24p states. The evolution of this mixed-parity wavepacket is characterized by time-dependent oscillations in the electric dipole moment, with a period of 2.9 ps. Approximately 5 ns after the wavepacket creation, a second 5 ns dye-laser promotes a second set of atoms from the $5p$ level into the $33s$ state. Because of the DD interaction, the second dye laser actually creates atom pairs whose electronic states are correlated via the resonant DD coupling. A $33s$+$34p$ wavepacket, oscillating with the same 2.9 ps period as the $25s$+$24p$ wavepacket, develops on the second set of atoms as a result of the correlation. A second, time-delayed ps THz pulse enables the detection of the coherent wavepacket motion on the two sets of atoms.

6.1 Introduction

For a pair of atoms A and B, as described in previous chapters, when the energy gap E from $|1$ to $|2$ on atom A is equal to the energy gap E' from $|2'$ to $|1'$ on atom B, resonant energy transfer will happen: $|1\ |1' \leftrightarrow |2\ |2'$ [1]. This process is also conveniently described in terms of collisions. In such a case, the composed eigenstate of AB pair is no longer $|1\ |1'$ or $|2\ |2'$. Instead dressed by dipole-dipole interactions, the atoms have eigenstates, in collision basis, which are linear combination of eigenstates in non-interacting basis: $|+ \ = (\ |1\ |1' \ + \ |2\ |2'\ )/\sqrt{2}$ and $| \quad = (\ |1\ |1' \quad |2\ |2'\ )/\sqrt{2}$. The state $|+$ has an eigenenergy $E+\epsilon$ and the state $|$ has an eigenenergy $E-\epsilon$, where ϵ is the amplitude of the DD interaction.

For a pair of atoms starting from $|1\ |1'$, the pair wavefunction can be written in the collision basis as $\Psi(t) = (\ |+ \ + \ | \quad e^{-i2\int \epsilon dt}\ )/\sqrt{2}$. The evolution of the wavefunction is clear. Now, we propose to consider another case. Suppose one atom in the pair is at first prepared in a coherent superposition state and the other one an eigenstate, the wavefunction of the pair is $\Psi(t) = (|1 \ + |2\ e^{-iEt})\ |1'\ /\sqrt{2}$. There is only one resonant transfer path $|1\ |\ 1' \leftrightarrow |2\ |\ 2'$. During DD interactions, the wavefunction is written as:

$$
\begin{aligned}
\Psi(t) &= (|+\ e^{-i(E+\epsilon)t} \ + | \quad e^{-i(E-\epsilon)t})/2 + |2\ |1'\ e^{-iEt}/\sqrt{2} \\
&= (|+ \ + | \quad e^{-i\ \varphi})/2 + |2\ |1'\ e^{-i\ \varphi/2}/\sqrt{2}
\end{aligned}
\tag{6.1}
$$

where $\varphi = 2\int \epsilon dt$ and it's called "collision phase". If φ is very small, when the resonant energy transfer is detuned, the wavefunction returns to non-interacting basis:

$$
\begin{aligned}
\Psi &\approx \frac{|1\ |1'}{\sqrt{2}} + \frac{i\ \varphi(|1\ |1' \quad |2\ |2'\ )}{2\sqrt{2}} + \frac{|2\ |1'}{\sqrt{2}} + \frac{i\ \varphi|2\ |1'}{2\sqrt{2}} \\
&\approx \frac{(1+i\ \varphi)(|1 \ + |2\ )|1'}{\sqrt{2}} \quad \frac{i\ \varphi|2\ (|2' \ + |1'\ )}{2\sqrt{2}}
\end{aligned}
\tag{6.2}
$$

Then the pair wavefunction evolution is:

$$\Psi(t) = \frac{(1 + i\ \ \varphi)(|1\ \ + |2\ e^{-iEt})|1'}{\sqrt{2}}\quad \frac{i\ \ \varphi|2\ (|2'\ \ + |1'\ e^{-iEt})}{2\sqrt{2}} \tag{6.3}$$

From equation (6.3), we see the DD interaction splits the coherent superposition state into two distinct pieces $(|1\ \ + |2\ e^{-iEt})|1'$ and $|2\ (|2'\ \ + |1'\ e^{-iEt})$. For the pair of atoms AB, these two combinations mean atom A is in an eigenstate with atom B in a coherent superposition state, or atom B is in an egenstate with atom A in a coherent superposition state. These two parts also exhibit time-dependent dipole moments oscillating with the same period and well-defined phase.

If the dipole-dipole interaction is long and the collision phase is large, the above derivation is not true. Our goal in this chapter is to reveal the phase shift between the original wavepacket of atom A and the induced wavepacket of atom B, when the collision phase is small. By doing this, we can demonstrate the transfer of coherence from atom A to atom B via DD interactions.

6.2 Experimental Procedure

A system that has been explored in detail in these papers [2,3] is utilized to achieve our goal. The resonant interaction is $25s33s \leftrightarrow 24p_{1/2}34p_{3/2}$. When the static offset field is about 3 V/cm, the energy gap between 25*s* and $24p_{1/2}$ is 11.4 cm^{-1}, which is the same as the gap between 33*s* and $34p_{3/2}$. In the experiment, cold Rb atoms are trapped in a MOT at state $5p_{3/2}$. A nanosecond dye laser pulse transfers one portion of atoms to 25*s*. These Rydberg atoms, considered as atoms A, are then transferred by a pulse of THz to a coherent state composed of 25s and 24*p* of roughly equal amount. Right after that, a second nanosecond dye laser pulse promotes another portion of $5p_{3/2}$ atoms to $33s$. These are atoms B. Now in the MOT, since atoms A and B are distributed randomly, the pair state of a lot of atoms satisfies our initial assumption: $(|25s \;\; + |24p \;)|33s \;$. The atoms are then interacting resonantly for a period *T* to generate another pair state $|24p \;\, (|34p \;\; + |33s \;)$. The coherent superposition of $33s$ and $34p$ will be probed by the second THz pulse. The first THz pulse which generates dipole moment oscillations on atoms A is scanned. Depending on the time between the two THz pulses or the "delay", amplitude transferred from $33s$ to $34p$ will interfere constructively or destructively with collisional excited amplitude, resulting in a modulation in the measured $34p$ population as a function of THz pulse delay [4]. After the second THz pulse, the resonant interaction will continue for a period *P*.

It is straightforward to think about showing the phase difference between $25s$ population oscillation and $34p$ population oscillation. But to detect $25s$, the required ionization filed is much larger than required for $34p$. For long DD interactions after the second THz pulse, the population of $34p$ is strongly modulated by 25s and there is no population oscillation phase difference between these two. This is easy to understand. In a long run, resonant energy transfer is saturated so that larger $25s$ population generates larger $34p$ population. So instead of comparing the phase between $25s$ and 34p, we consider about another convenient method. The phase of $34p$ population oscillation at a short interaction is compared with the phase of $34p$ population oscillation at a long interaction (which is synchronized with $25s$ population oscillation). In an other word, we just need to get the phase difference of $34p$ population oscillations at a small *P* and a large *P*. At last an ionization field ramp is used to detect the amplitude of $34p$. In principle, the phase difference of $33s$ population oscillations is also able to demonstrate a coherence transfer, but the population oscillation of $33s$ is not as good as that of $34p$ in experiment. So we stick to detecting the population oscillation of $34p$.

Above discussion provides an idea about the experiment procedure. The next step is to determine the proper values of T and P in the experiment. On one hand, T should be long enough for detectable $34p$ population. On the other hand, if T is too large, collision phase φ will be too large and the approximation is not satisfied. Our estimate of T for a MOT with Rydberg density ~$3\times 10^9\mathrm{cm}^{-1}$ is from 10s of ns to 100 ns (refer to Figure 6.1). In practice, we find 90 ns gives us a pretty good result.

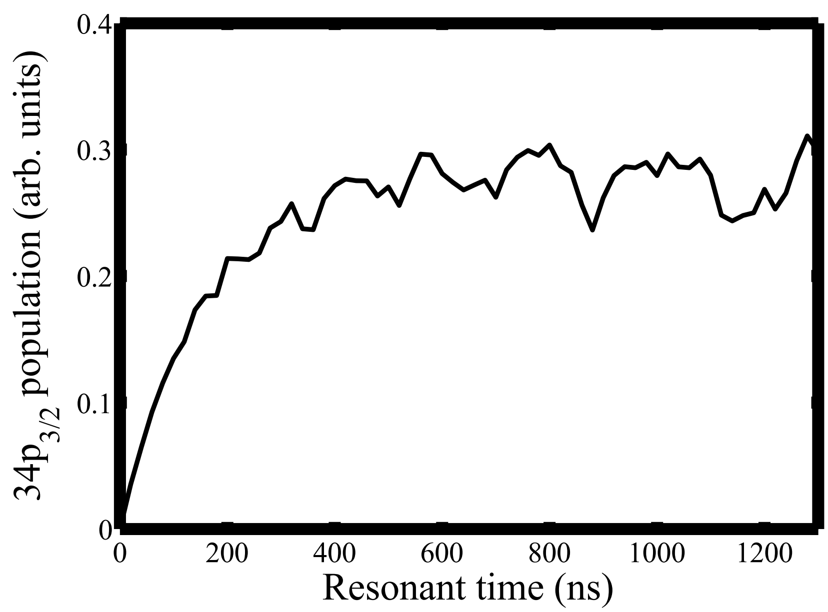

Figure 6.1: Calculated generation of $34p$ as a function of interaction time T shows how we estimate the proper T. The initial condition is that half of atoms in state $25s$ and other half state $33s$. After about 300 ns, the transfer is saturated. In practice, for a T smaller than 90 ns, it is difficult to detect the amplitude interference under the current experiment condition.

P is the time after the second THz pulse. When P is small, the assumption of small collision phase is satisfied and it is expected to get a phase difference between the oscillations of dipole moments of atoms A and atoms B. When P is large, the assumption is not satisfied and the phase difference is minimized. To show such a change, *P* needs to be extreme values. In practice, to be small, *P* is set to be as close as possible to 0. Due to setup limit, the real value is estimated to be ~3 ns. For big *P* values, more than 300 ns is enough to synchronize the population oscillations of 25*s* and 34*p*. In the experiment, the value set for large *P* is 600 nanoseconds.

6.3 Experimental Results

The population oscillation plot of different states is achieved by scanning the delay between the two THz pulses. Figure 6.2 shows the amplitude oscillation of 34*p* at different *P*. The phase difference between short delay oscillation and long delay oscillation of 34*p* population is positive. 25*s* population oscillation is synchronized with 34*p* population oscillation at large *P* and 180 degrees different from the 24*p* population oscillation. So the phase difference between the original 24*p* population oscillation and the induced 34*p* population oscillation is positive.

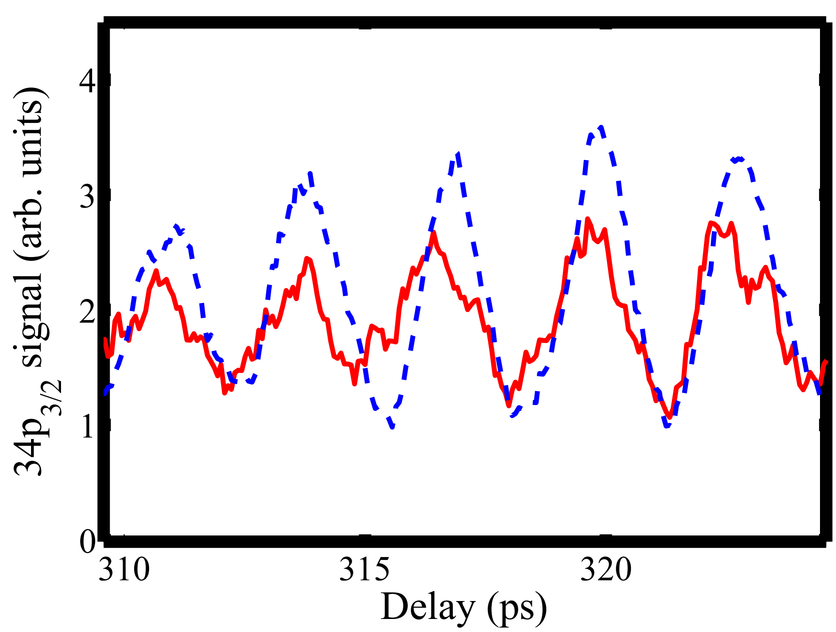

Figure 6.2: 34*p* population oscillation at different *P*. The red solid line represents the 34*p* signal when *P* is around 0 ns. The blue dash line represents the 34*p* signal when *P* is

around 600 ns. The measured signals are very noisy but from this snippet we can tell that the red oscillation has a phase advanced than the blue oscillation.

The next step is to get quantified values of the phase shift. To look into details, the collected signal is not only investigated in the temporal domain but also in the frequency domain. Figure 6.3 shows the signal converted into frequency domain using fast Fourier transfer method. The phase is measured at wavenumber 11.4 cm^{-1} to get the result. The phase difference of dipole moment oscillation for short P and long P is 21 degrees for the setup discussed above.

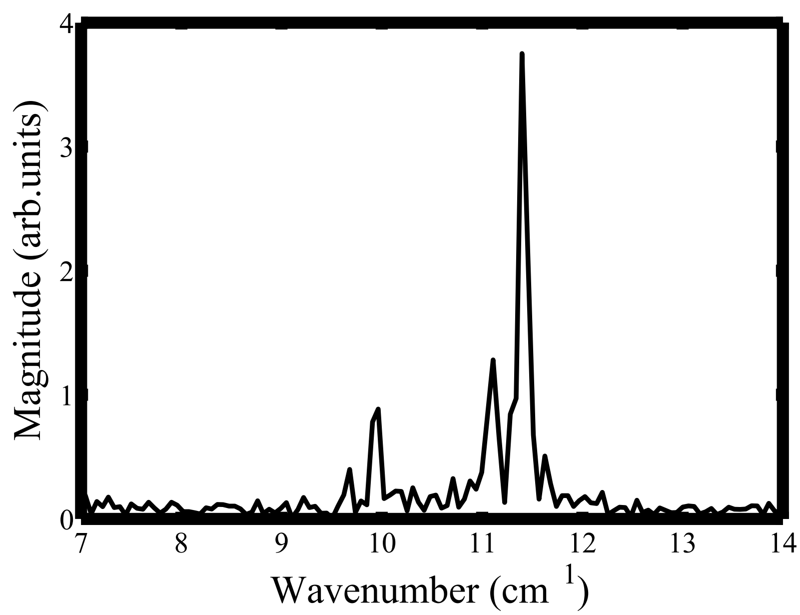

Figure 6.3: Fast Fourier transfer result of $34p$ population oscillation in the temporal domain when P is large. (The FFT is similar when P is short.) There are three observable peaks from this plot. 11.4 cm^{-1} corresponds to the transfer between $25s$ to $24p_{1/2}$ or $33s$ to $34p_{3/2}$ and the phase at this peak is what we are interested in. 11.1 cm^{-1} corresponds to the transfer from $25s$ to 24 $p_{3/2}$ and 9.9 cm^{-1} $25s$ to $25p_{3/2}$. Those peaks are not what we are interested in, but they do provide us one way to filter out bad measured runs since the phase at those frequencies should keep relatively consistent between different runs.

6.4 Discussion

The result 21 degrees is smaller than what we expected in the first place. From our simulation, a value close to 90 degrees is expected. In this discussion, the causes of the result being smaller than expectation is discussed.

The dipole-dipole interaction has to be small to make Equation (6.3) a reasonable derivation. But in experiment, larger DD interactions provides better measurements of 34*p* population and its oscillation. We have to balance between these two opposite choices and find a proper DD interaction time for our system. It's reasonable that the result from practical compromised choice is not as good as the ideal calculation.

Simulation to calculate the phase difference has been done for different P. This simulation is pretty rough because what the THz pulses look like or what their amplitudes are is not exactly known. Different combinations of parameters have been tried to build THz pulses so that the population redistribution by THz pulses is as close to the experiment observation as possible. As shown in Figure 6.4, the phase between 34*p* oscillation and 25*s* oscillation quickly declines to 0. Ideally, P should be very close to 0. In the experiment, we estimate the real P to be around 3ns or even bigger due to issues such as instability of synchronized system and measurement limit. When P is not 0, the result is shifted from the ideal expectation.

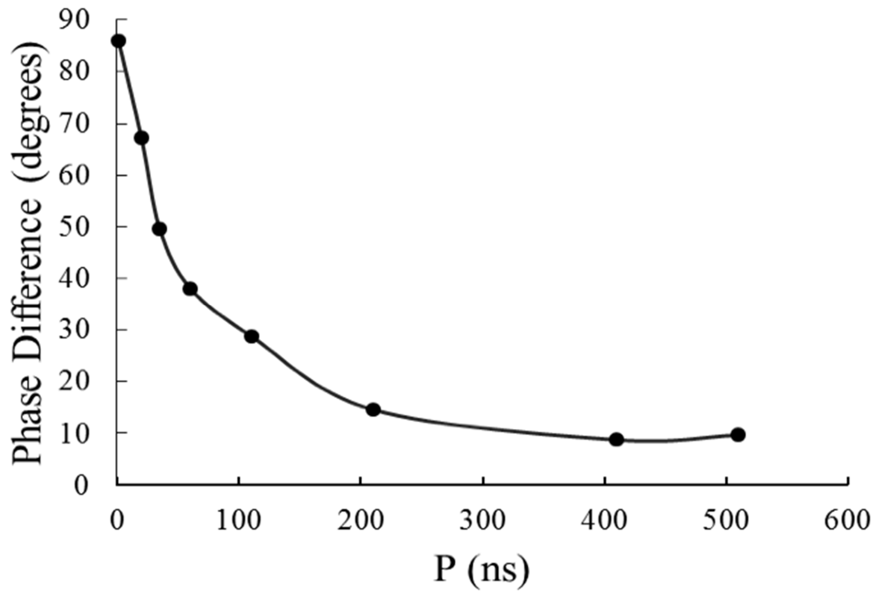

Figure 6.4: Calculated phase difference between 34*p* and 25*s* dipole moment oscillation as a function of period *P*. The phase difference declines pretty quickly.

In experiment, such a decline is even faster. We can not detect distinguishable phase difference when *P* is larger than 10 ns. That's why we only compare the results when *P* is around 0 and when *P* is very large.

6.5 Conclusion

In theory, we have explored the mechanism of coherence transfer of Rydberg wavepackets from one atom to a nearby atom via DD interactions. In experiment, we have revealed the coherence transfer by detecting the phase difference between the original wavepacket and the induced wavepacket. The detected phases difference between induced 34*p* population oscillation and the original 25*s* population oscillation is 21 degrees, which is smaller than the expected 90 degrees. Through calculation and analysis, we have explained that the causes of the error are setup limits and the nature of the fast decay of the phase difference.

Bibliography

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