Suppression of Magnetic State Decoherence Using Ultrafast Optical Pulses

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(March 31, 2022)

32.80.Qk, 34.50.Rk, 34.20.Cf

It is shown that the magnetic state decoherence produced by collisions in a thermal vapor can be suppressed by the application of a train of ultrafast optical pulses.

In a beautiful experiment, Itano et al. demonstrated the Quantum Zeno effect [1]. A radio frequency pi pulse having a duration on the order of 250 ms was applied to a ground state hyperfine transition. At the same time, a series of radiation pulses was used to drive a strongly coupled ground to excited state uv transition. The rf and strong transitions shared the same ground state level. Itano et al. showed that excitation of the rf transition could be suppressed by the uv pulses. They interpreted the result in terms of collapse of the wave function - spontaneous emission from the excited state during the uv pulses is a signature that the uv pulse projected the atom into its ground state; the lack of such spontaneous emission implies projection into the final state of the rf transition. This paper triggered a great deal of discussion, especially with regards to the interpretation of the results [2].

A necessary condition for a quantum Zeno effect is a perturbation of a state amplitude on a time scale that is short compared with the correlation time of the process inducing the transition. In the experiment of Itano et al., this time scale is simply the duration of the pi pulse, 256 ms. On the other hand, if one wished to inhibit particle decay or spontaneous emission [3], it would be necessary to apply perturbations on a time scale that is short compared with the correlation time of the vacuum, an all but impossible task. In this paper, we consider the inhibition of collisional, magnetic state decoherence, by the application of a train of ultrafast, optical pulses. This correlation time of the collisional perturbations resulting in magnetic state decoherence is of order of the duration of a collision and is intermediate between that for the coherent pi pulse applied by Itano et al. and the almost instantaneous, quantum jump-like process produced by the vacuum field. It should be noted that related schemes have been proposed for inhibiting decoherence in systems involving quantum computation [4], but the spirit of these proposals differs markedly from the one presented herein.

The rapid perturbations of the system are a necessary, but not sufficient, condition for a mechanism to qualify as a Quantum Zeno effect. The perturbations must involve some "measurement" on the system for the "Quantum Zeno" label to apply. The suppression of magnetic state coherence discussed in this paper is not a Quantum Zeno effect in this sense. We will return to this point below.

We envision an experiment in which "active atoms" in a thermal vapor undergo collisions with a bath of foreign gas perturbers. A possible level scheme for the active atoms is depicted in Fig. 1. At some initial time, an ultrashort pulse excites an atom from its ground state, having angular momentum \( J = 0 \), to the \( m = 0 \) sublevel of an excited state having \( J = 1 \). The duration of the excitation pulse \( \tau_p \) is much shorter than the duration of a collision \( \tau_c \) (\( \tau_c \) is typically of order 1 ps). As a result of elastic collisions with the ground state perturbers, population in the \( J = 1 \) sublevels equilibrate at a rate \( \Gamma_{col} \) that is typically of order \( 10^7 - 10^8 \) s\(^{-1} \) per Torr of perturber pressure. The transfer to the \( m = 1 \) substate is probed by a circularly polarized pulse acting on the \( J = 1, m = 0 \rightarrow J = 0 \) excited state transition, applied at a time \( \Gamma_{col}^{-1} \) following the initial excitation pulse. For the sake of definiteness, we assume that the perturber pressure is such that equilibration occurs in a time \( \Gamma_{col}^{-1} = 0.1 - 1.0 \) ns. The question that we address in this paper is "How can one inhibit this magnetic state decoherence by subjecting the active atoms to additional external radiation fields?"

![FIG. 1. Energy level diagram. The collisional interaction couples the magnetic sublevels in the \( J = 1 \) state.](image-url)
As was mentioned above, the key to any Zeno-type effect is to disrupt the coherent evolution a system from its initial to final state. In our case, the coherent evolution from the initial m = 0 states to the final m = ±1 states is driven by the collisional interaction. Thus it is necessary to disturb the system on a time scale that is short compared with the collision duration \( \tau_c \). To do this, we apply a continuous train of ultrashort pulses that couple the \( m = 0 \) level to the excited state having \( J = 0 \) shown in Fig. 1. The pulses are assumed to have durations \( \tau_p \ll \tau_c \) and are assumed to be off-resonance; that is, the atom-field detuning is large compared with \( \tau_p^{-1} \). As such, each pulse simply produces an ac Stark shift of the \( m = 0 \) sublevel of the \( J = 1 \) state, resulting in a phase shift of this state amplitude. As a consequence, the external pulses break the collision-induced, coherent evolution of the atom from its initial \( m = 0 \) state to the \( m = \pm 1 \) states. If the pulse strengths are chosen such that the phase shift is a random number, modulo \( 2\pi \), and if many pulses occur during the collision duration \( \tau_c \), then the atom will be frozen in its initial state. It is interesting to note that collisions, which are normally viewed as a decohering process, must be viewed as a coherent driving mechanism on the time scales considered in this work.

To obtain a qualitative understanding of this effect, it is sufficient to consider a model, two-level system, consisting of an initial state \( |0\rangle \) (corresponding to the \( J = 1 \), \( m = 0 \) state) and a final state \( |1\rangle \) (corresponding to the \( J = 1 \), \( m = 1 \) state, for example). The Hamiltonian for this two-state system is taken as

\[
H = V_c(t) \langle 0 | 1 \rangle + |1 \rangle \langle 0 | + \hbar \sum_i \Delta_s(t_i) \tau_p \delta(t-t_i) |0 \rangle \langle 0 |,
\]

where \( V_c(t) \) is a collisional perturbation that couples the two, degenerate states, and \( \Delta_s(t_i) \) is the ac Stark shift of state \( |0\rangle \) produced by the external pulse occurring at \( t = t_i \). For simplicity, we take \( V_c(t) \) to be a square pulse, \( V_c(t, b) = \hbar \beta(b) \), for \( 0 \leq t \leq \tau_c \). The quantity \( b \) is the impact parameter of the collision. Without loss of generality, we can take the collision to start at \( t = 0 \). The collision duration \( \tau_c \) can be written in terms of the impact parameter \( b \) characterizing the collision and the relative active atom-perturber speed \( u \) as \( \tau_c(b) = b/u \).

Moreover, to simulate a van der Waals interaction, we set \( \beta(b) = (C/b_0^6) (b/b_0)^6 \), where \( C \) and \( b_0 \) are constants chosen such that \( 2C/(b_0^6u) = 1 \). The quantity \( b_0 \) is an effective Weikspop radius for this problem. An average over \( b \) will be taken to calculate the transition rate.

The external pulse train is modeled in two ways. In model A, the pulses occur at random times with some average separation \( T \) between the pulses. In model B, the pulses are evenly spaced with separation \( T \). In both models, the pulse areas \( \Delta_s(t_i) \tau_p \) are taken to be random numbers between 0 and \( 2\pi \). A quantity of importance is the average number of pulses, \( n_0 = \tau_c(b_0)/T = b_0/(uT) \), for a collision having impact parameter \( b_0 \).

### A. Randomly-spaced pulses

The randomly spaced, radiative pulses act on this two-level system in a manner analogous to the way collisions modify atomic electronic-state coherence. In other words, the pulses do not affect the state populations, but do modify the coherence between the levels. The pulses can be treated in an impact approximation, such that during a collision, the time rate of change of density matrix elements resulting from the pulses is \( \dot{\rho}_{00} = \dot{\rho}_{11} = 0 \) and

\[
\dot{\rho}_{10}/\rho_{10} = \dot{\rho}_{01}/\rho_{01} = -\Gamma \left( 1 - e^{-i\Delta_s(t_i)/\tau_p} \right) = -\Gamma,
\]

where \( \Gamma = T^{-1} \) is the average pulse rate and we have used the fact that the pulse area is a random number between 0 and \( 2\pi \). Taking into account the collisional coupling \( V_c(t, b) \) between the levels, one obtains evolution equations for components of the Bloch vector \( w = \rho_{11} - \rho_{00} = 2\rho_{11} - 1 \), \( v = i(\rho_{01} - \rho_{10}) \) as

\[
dw/dx = U(y)v, \quad dv/dx = -U(y)w - n(y)v,
\]

where \( x = t/\tau_c(b) \) is a dimensionless time, \( y = b/b_0 \) is a relative impact parameter, and \( U(y) = y^{-5} \) and \( n(y) = n_0 y \) are dimensionless frequencies. These equations are solved subject to the initial condition \( w(0) = -1; v(0) = 0 \), to obtain the value \( \rho_{11}(x = 1, y, n_0) = \left[w(x = 1, y) + 1\right]/2 \). The relative transition rate \( S \) is given by

\[
S(n_0) = 2\pi N u b_0^2 \int_0^\infty y dy \rho_{11}(x = 1, y, n_0)/2,
\]

where \( N \) is the perturber density. A coefficient, \( R(n_0) \), which measures the suppression of decoherence, can be defined as

\[
R(n_0) = \int_0^\infty y dy \rho_{11}(x = 1, y, n_0)/\int_0^\infty y dy \rho_{11}(x = 1, y, 0)
\]

Solving Eqs. (3), one finds

\[
\rho_{11}(x = 1, y, n_0) = \left[1 - \frac{r_1}{r_2} - \frac{r_2}{r_1} \left(e^{-r_1} - e^{-r_2}\right)\right]/2;
\]

\[
r_{1,2} = \left(-n_0 y \pm \sqrt{(n_0 y)^2 - 4y^{-10}}\right)/2.
\]

It is now an easy matter to numerically integrate Eqs. (2) to obtain \( R(n_0) \). Before presenting the numerical results, we can look at some limiting cases which provide insight into the physical origin of the suppression of decoherence.

A plot of \( \rho_{11}(x = 1, y, n_0) \) as a function of \( y = b/b_0 \) is shown in Fig. 1 for several values of \( n_0 \). With decreasing \( y \), \( \rho_{11} \) increases monotonically to some maximum value \( \rho_{11}(y_m) \) and then begins to oscillate about \( \rho_{11} = 1/2 \).
with increasing amplitude. One concludes from such plots that two effects contribute to the suppression of coherence. The first effect, important for large \( n_0 \), is a reduction in the value of \( \rho_{11}(y_m) \). The second effect, important for \( n_0 \) of order unity, is a decrease in the value of \( \rho_{11}(y_m) \). Let us examine these two effects separately.

For very large \( n_0 \), \( n_0^{5/6} \gg 1 \), one can approximate \( \rho_{11} \) over the range of \( y \) contributing significantly to the integral \( \rho_{11} \) as \( \rho_{11}(x = 1, y, n_0) \sim \left( 1 - e^{-y^{-11/n_0}} \right)/2 \). By evaluating the integrals in (5), one finds a suppression of decoherence ratio given by

\[
R(n_0) = 0.95/n_{0}^{2/11}. \tag{7}
\]

The \( n_{0}^{-2/11} \) dependence is a general result for a collisional interaction that varies as the interatomic separation to the minus 6th power. It can be understood rather easily. The pulses break up the collision into \( n_0 y \) segments, each having a (dimensionless) time duration \( x_b = 1/(n_0 y) \). Each segment provides a perturbative contribution to \( \rho_{11} \) of order \( y^{-10(n_0 y)^{-2}} \), provided \( y < y_w \), where \( y_w \) is to be determined below. The total population from the entire collision interval varies as \( \rho_{11} \sim y^{-10(n_0 y)^{-2}} n_0 y = y^{-11/n_0} \). Of course, \( \rho_{11} \) cannot exceed unity. One can define an effective relative Weisskopf radius, \( y_w \), as one for which \( \rho_{11} = 1 \), namely \( y_w = b/b_0 = n_{0}^{-1/11} \). The total transition rate varies as \( y_w^2 \sim n_{0}^{-2/11} \), in agreement with (5). As \( n_0 \rightarrow \infty \), the atom is frozen in its initial state.

For values of \( n_0 \) of order unity, the dominant cause of the suppression of decoherence is a decrease in the value of \( \rho_{11}(y_m) \), rather than the relatively small decrease in \( y_m \) from its value when \( n_0 = 0 \). For values \( n_0 \leq 3 \), approximately 45% of the contribution to the transition rate \( S(n_0) \) originates from \( y > y_m \), and, for these values of \( n_0 \), \( y_m \sim \pi^{-1/5} \) and \( \rho_{11}(y_m) \sim 1 + e^{-n_0/2\pi^{1/5}}/2 \). This allows us to estimate the suppression of decoherence ratio as \( R(n_0) = (0.55 + 4.5(1 + e^{-n_0/2\pi^{1/5}}))/2 \), such that \( R(1) = 0.93, R(2) = 0.88, R(3) = 0.84 \). These values are approximately 70% of the corresponding numerical results, indicating that the decrease in \( \rho_{11}(y_m) \) accounts for approximately 70% of the suppression at low \( n_0 \), with the remaining 30% coming from a decrease in \( y_m \). The first few collisions are relatively efficient in suppressing decoherence. With increasing \( n_0 \), the suppression process slows, varying as \( n_0^{-2/11} \). In Fig. 3, the suppression of decoherence ratio \( R(n_0) \), obtained by a numerical solution of Eq. (5), is plotted as a function of \( n_0 \).

![Graph of the suppression of decoherence ratio \( R \) as a function of \( n_0 \) for randomly and uniformly spaced pulses.](image)

**B. Uniformly Spaced Pulses**

We consider now the case of equally spaced pulses, having effective pulse areas that are randomly chosen, modulo \( 2\pi \). The time between pulses is \( T \), and \( n_0 = \tau_c(b_0)/T \). For a relative impact parameter \( y = b/b_0 \), with \( m \leq n(y) = n_0 y \leq m + 1 \), where \( m \) is a positive integer or zero, exactly \( m \) or \( m + 1 \) pulses occur. The effect of the pulses is calculated easily using the Bloch vector. At \( x = 0, w = -1 \) and \( v = 0 \). The Bloch vector then undergoes free evolution at frequency \( U(y) = y^{-5} \) up until the (dimensionless) time of the first pulse, \( x_s = t_s/\tau_c(b) \). The pulse randomizes the phase of the Bloch vector, so that the average Bloch vector following the pulse is projected onto the \( w \) axis. From \( x = x_s \), \( x_s + T/\tau_c(b) = x_s + 1/n(y) \), the Bloch vector again precesses freely and acquires a phase \( UT = y^{-5} n(y) = y^{-6}/n_0 \), at which time the next pulse projects it back onto the \( w \) axis. Taking into account the periods of free precession and projection, and averaging over the time \( x_s \) at which the first pulse occurs, one finds

\[
w(y) = [1 - n(y)] \cos[y^{-5}] + n(y) \int_0^1 dx_s \cos[y^{-5} x_s] \cos[y^{-5}(1 - x_s)];
\]

\[0 \leq y \leq 1/n_0,\]
w(y) = \left[ m + 1 - n(y) \right] \left[ \left( m + 1 \right) / n(y) - 1 \right]^{-1} \\
\times \int_{1-n/y}^{1/n(y)} dx_s \cos[y^{-5}x_s] \cos[n^m[y^{-6}/n_0]] \\
\times \cos[y^{-5}(1 - x_s - (m - 1)/n(y))] \\
+ [n(y) - m] [1 - m/n(y)]^{-1} \\
\times \int_{0}^{1-m/n(y)} dx_s \cos[y^{-5}x_s] \cos[m[y^{-6}/n_0]] \\
\times \cos[y^{-5}(1 - x_s - m/n(y))]; \\
\text{for } m/n_0 \leq y \leq (m + 1)/n_0 \text{ for } m \geq 1. \tag{8}

In the limit that \( n_0 > 1 \), for all impact parameters that contribute significantly to the transition rate, approximately \( n(y) \) pulses occur at relative impact parameter \( y \), implying that \( w(y) \sim \cos^{n(y)\{y^{-5}/n(y)\}} \) and

\[
R(n_0) = \frac{\langle 1 - \cos^{n_0 y}\{y^{-6}/n_0\} \rangle}{\langle 1 - \cos\{y^{-5}\} \rangle} \\
\sim \frac{\langle 1 - \left( 1 - y^{-12}/2n_0^2\{n_0 y\} \right) \rangle}{\langle 1 - \cos\{y^{-5}\} \rangle} \\
\sim \frac{\langle 1 - e^{-y^{-11}/2n_0} \rangle}{\langle 1 - \cos\{y^{-5}\} \rangle} = \frac{0.84}{n_0^{2/11}},
\]

which is the same functional dependence found for the randomly spaced pulses. Note that the form \( \{ 1 - \cos^{n(y)}\{y^{-15}/n(y)\} \} \) is identical to that found in theories of the Zeno effect [5].

The suppression of decoherence ratio \( R(n_0) \), obtained from Eqs. (4) and (8) [using \( \rho_{11} = (1 + w)/2 \)], is plotted in Fig. 3. The fact that it lies below that for randomly spaced pulses is connected with the difference in the average collisional phase shift acquired between radiation pulses for the two models. The oscillations in \( R(n_0) \) appear to be an artifact of our square pulse collision model. In the absence of the pulses, the first maximum in the transition cross section occurs for \( y_{max} = (\pi)^{-1/5} \), corresponding to a \( \pi \) collision pulse. With increasing \( n_0 \), the pulses divide the collision duration into approximately \( n(y) \) equal intervals. If these pulse intervals are odd or even multiples of \( \pi \), one can enhance or suppress the contribution to the transition rate at specific impact parameters. Numerical calculations carried out for a smooth interatomic potential do not exhibit these oscillations.

### C. Discussion

Although the collisional interaction has been modeled as a square pulse, the qualitative nature of the results is unchanged for a more realistic collisional interaction, including level shifts. In fact, for a smooth interatomic potential that allows for an increased number of radiation pulses over the duration of the collisional interaction, the suppression is slightly enhanced from the square pulse values. Although the pulses are assumed to drive only the \( J = 1, m = 0 \rightarrow J = 0 \), excited state transition, it is necessary only that the incident pulses produce different phase shifts on the \( J = 1, m = 0 \) and \( J = 1, m = 1 \) state amplitudes.

To observe the suppression of decoherence, one could use Yb as the active atom and Xe perturbers. The Weiskopf radius for magnetic decoherence is about 1.0 nm \( \Box \), yielding a decoherence rate of \( \simeq 10^{10} \text{s}^{-1} \) at 500 Torr of Xe pressure at 300°C, and a collision duration \( \tau_c(b_0) \simeq 2.5 \mu s \). Thus, by choosing a pulse train having pulses of duration \( \tau_p = 100 \text{fs} \), separated by 0.5 ps, it is possible to have 5 pulses per collision. If an experiment is carried out with an overall time of 100 ps (time from initial excitation to probing of the final state), one needs a train of about 200 pulses. To achieve a phase shift \( \Delta \sigma \tau_p \) of order \( 2\pi \) and maintain adiabaticity, one can take the detuning \( \delta = 3 \times 10^{13} \text{s}^{-1} \) and the Rabi frequency \( \Omega \simeq 1 \times 10^{14} \text{s}^{-1} \) on the \( J = 1, m = 0 \rightarrow J = 0 \), excited state transition \( \Box \). The corresponding, power density is \( \simeq 1.5 \times 10^{11} \text{W/cm}^2 \), and the power per pulse is \( \simeq 150 \text{mJ} \) (assuming a 1 mm\(^2\) focal spot size). This is a rather modest power requirement. With 5 pulses/collision duration, one can expect a relative suppression of magnetic state decoherence of order 40%.

Finally, we should like to comment on whether or not the effect described in this work constitutes a Quantum Zeno effect. Normally, the Quantum Zeno effect is presented as a projection of a quantum system onto a given state as a result of a measurement on the system. In the experiment of Itano et al., this "measurement" is reflected by the presence or absence of spontaneously emitted radiation during each \( w \) "measurement" pulse. The measurement pulse must be sufficiently long to produce a high likelihood of spontaneous emission whenever the atom is "projected" into the initial state by the pulse. Following each measurement pulse, the off-diagonal density matrix element for the two states of the \( rf \) transition goes to zero. In our experiment involving off-resonant pulses, the number of Rayleigh photons scattered from the \( J = 0 \) level during each applied pulse is much less than unity. As such, there is no Quantum Zeno effect, even if suppression of magnetic state decoherence occurs. On average, each pulse having random area destroys the coherence between the \( J = 1, m = 0 \) and \( J = 1, m = \pm 1 \) state amplitudes, but does not kill this coherence for a single atom. With an increasing number of radiation pulses, \( n_0 \), however, both the average value and the standard deviation of the transition probability tends to zero as \( n_0^{-1} \) for each atom in the ensemble.

The experiment of Itano et al. could be modified to allow for a comparison with the theory presented herein, and to observe the transition into the Quantum Zeno regime. If the pulses that drive the strong transition are replaced by a sequence of off-resonant pulses, each pulse having a duration \( \tau_p \) much less than the time, \( T_{\pi} \), required for the pi pulse to drive the weak transition, and each pulse having an effective area, \( \Delta_s \tau_p = (\Omega^2/4\delta)\tau_p \), that is random in the domain \([0,2\pi]\), then the pulses will...
suppress the excitation of the weak transition (it is assumed that $\Omega/\delta \ll 1$). If the upper state decay rate is $\gamma$, then the average number of Rayleigh photons scattered during each pulse is $n = (\Omega/4\delta)^2 \gamma \tau_p$. For $n < 1$, there is suppression of the transition rate as in our case, while, for $n \gtrsim 1$, there is suppression and a Quantum Zeno effect.

There is no average over impact parameter, since exactly $[T_\pi/T]$ or $([T_\pi/T]+1)$ pulses in each interval between the pulses, where $[x]$ indicates the integer part of $x$.

### D. Acknowledgments

PRB is pleased to acknowledge helpful discussions with R. Merlin, A. Rojo and J. Thomas. This research is supported by the National Science Foundation under grant PHY-9800981 and by the U. S. Army Research Office under grants DAAG55-97-0113 and DAAH04-96-0160.

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