

## Suppression of Magnetic State Decoherence Using Ultrafast Optical Pulses

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

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The subject of decoherence has attracted a great deal of attention in the past few years. In atomic vapors, one source of *magnetic state decoherence* is collisions, since collisions redistribute the population among different Zeeman sublevels. The question that we address in this paper is “How can one inhibit this magnetic state decoherence by subjecting the atoms to additional external radiation fields?” The key to suppressing decohering transitions produced by a thermal bath is to perturb the relevant state amplitudes on a time scale that is *short compared with the correlation time of the bath*. Thus, to suppress spontaneous decay, the decaying particle must be perturbed on a time scale that is shorter than the correlation time of the vacuum field, an all but impossible task. On the other hand, the correlation time of the collisional perturbations that produce magnetic state decoherence is on the order of the collision duration, typically of order 1 ps. It is possible to apply several ultrafast optical pulses during a *single* collision in a manner that inhibits magnetic state decoherence. As a consequence, one has a practical means for preserving magnetic state coherence in the presence of collisions. It should be noted that related schemes have been proposed for inhibiting decoherence in systems involving quantum computation and control [1]. The relationship of suppression of magnetic state decoherence to the quantum Zeno effect [2–4] is discussed at the end of the paper.

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 is much shorter than the collision duration  $\tau_c$ . As a result of elastic collisions with the ground state perturbers, population in the  $J = 1$  sublevels equilibrate at a rate  $\Gamma_{\text{col}}$  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  $J = 1, m = 1 \rightarrow J = 0$  excited state transition, applied at a time  $\Gamma_{\text{col}}^{-1}$  following the initial excitation pulse. For the sake of definiteness, we assume that the perturber pressure is such that equilibration occurs in a time  $\Gamma_{\text{col}}^{-1} = 0.1$ – $1.0$  ns. Spontaneous emission is neglected on this time scale.

As was mentioned above, one must disrupt the coherent evolution of a system from its initial to final state. In our case, the coherent evolution from the initial  $m = 0$  state to the final  $m = \pm 1$  states is driven by the collisional interaction. Thus it is necessary to perturb 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 duration  $\tau_p \ll \tau_c$  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. The pulse strengths can be chosen in a deterministic way so as to minimize the transition probability [1]. Rather than follow this approach, we assume that the pulse strengths are chosen such that each phase shift is a random number, mod  $2\pi$ . In this way, the pulse train acts as an irreversible bath. If many pulses occur during the collision duration  $\tau_c$ , the atom is 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  $J = 1, m = 0$

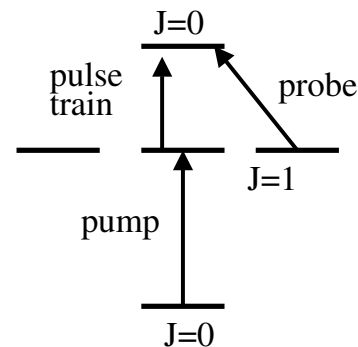


FIG. 1. Energy level diagram. The collisional interaction couples the magnetic sublevels in the  $J = 1$  state.

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

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

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  $|0\rangle$ , proportional to the intensity of 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. 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  $\beta(b) = (C/b_0^6)(b_0/b)^6$ , where  $C$  and  $b_0$  are constants chosen such that  $2C/(b_0^5 u) = 1$ . The quantity  $b_0$  is an effective Weisskopf 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

$$\dot{\rho}_{10}/\rho_{10} = \dot{\rho}_{01}/\rho_{01} = -\Gamma \langle 1 - e^{-i\Delta_s(t_i)\tau_p} \rangle = -\Gamma, \quad (2)$$

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  $w = \rho_{11} - \rho_{00} = 2\rho_{11} - 1$ ,  $v = i(\rho_{10} - \rho_{01})$  as

$$dw/dx = U(y)v, \quad dv/dx = -U(y)w - n(y)v, \quad (3)$$

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  $\rho_{11}(x = 1, y, n_0) = [w(x = 1, y) + 1]/2$ . The relative transition rate  $S$  is given by

$$S(n_0) = 2\pi N u b_0^2 \int_0^\infty y dy \rho_{11}(x = 1, y, n_0), \quad (4)$$

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

$$R(n_0) = \int_0^\infty y dy \rho_{11}(x = 1, y, n_0) / \int_0^\infty y dy \rho_{11}(x = 1, y, 0). \quad (5)$$

Solving Eqs. (3), one finds

$$\rho_{11}(x = 1, y, n_0) = \left[ 1 - \frac{r_2}{r_2 - r_1} \left( e^{r_1} - \frac{r_1}{r_2} e^{r_2} \right) \right] / 2; \quad (6a)$$

$$r_{1,2} = \left( -n_0 y \pm \sqrt{(n_0 y)^2 - 4y^{-10}} \right) / 2. \quad (6b)$$

It is now an easy matter to numerically integrate Eqs. (5) 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. 2 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  $y_m$ . 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  $n_0$ ,  $n_0^{5/66} \gg 1$ , one can approximate  $\rho_{11}$  over the range of  $y$  contributing significantly to the integral (4) as  $\rho_{11}(x = 1, y, n_0) \sim (1 - e^{-y^{-11}/n_0})/2$ . By evaluating the integrals in (5), one finds a suppression of decoherence ratio given by

$$R(n_0) = 0.95/n_0^{2/11}. \quad (7)$$

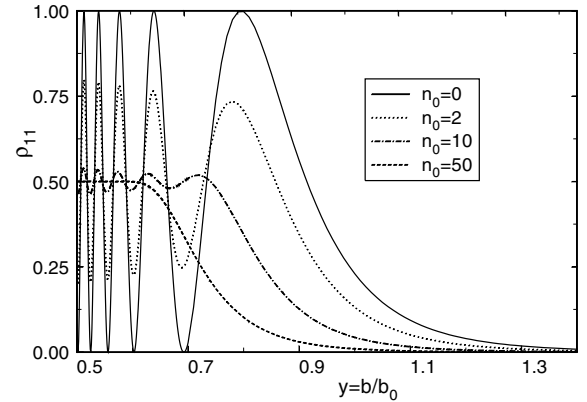


FIG. 2. Graph of  $\rho_{11}$  as a function of  $y = b/b_0$  for several values of  $n_0$ . For values  $0 \leq y \leq 0.45$  not shown on the graph,  $\rho_{11}$  oscillates about an average value of  $1/2$ . For  $n_0 \neq 0$ , the oscillation amplitude increases with decreasing  $y$ .

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, on average. Since the relative phase changes randomly with each radiation pulse, the final state probability amplitude,  $a_1(t)$ , undergoes a random walk, each of whose segments has a length  $(1/2)y^{-5}x_r$ , where  $x_r$  is the (dimensionless) duration of a single step [ $\langle x_r \rangle = (n_0 y)^{-1}$ ,  $\langle x_r^2 \rangle = 2(n_0 y)^{-2}$ ]. As a consequence, after  $n_0 y$  steps,  $\rho_{11} = \langle |a_1(t)|^2 \rangle \sim (y^{-10}/4)[2(n_0 y)^{-2}](n_0 y) = y^{-11}/2n_0$ . 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  $y_w = b_w/b_0 = (2n_0)^{-1/11}$ . The total transition rate varies as  $y_w^2 \sim n_0^{-2/11}$ , in agreement with (7). As  $n_0 \sim \infty$ , 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  $R(n_0) = [0.55 + 0.45(1 + e^{-n_0/2\pi^{1/5}})]/2$ , 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  $n_0$ .

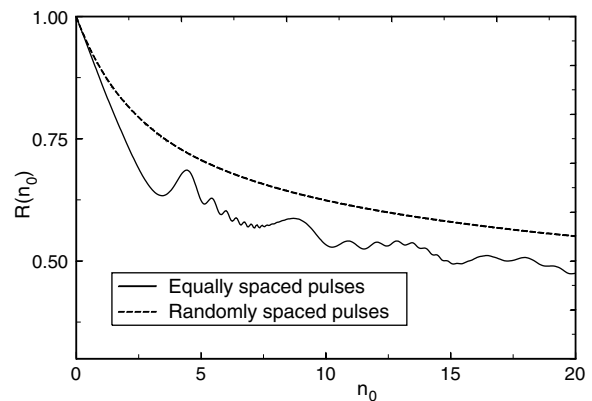


FIG. 3. Graph of the suppression of decoherence ratio  $R$  as a function of  $n_0$  for randomly and uniformly spaced pulses.

**B. Uniformly Spaced Pulses.**—We consider now the case of equally spaced pulses, having effective pulse areas that are randomly chosen, mod  $2\pi$ . 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  $x = x_s$  to  $x_s + T/\tau_c(b) = x_s + 1/n(y)$ , the Bloch vector again precesses freely and acquires a phase  $UT/\tau_c(b) = y^{-5}/n(y) = y^{-6}/n_0$ , 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

$$\begin{aligned}
 w(y) &= [1 - n(y)] \cos[y^{-5}] + n(y) \int_0^1 dx_s \cos[y^{-5}x_s] \cos[y^{-5}(1 - x_s)]; & 0 \leq y \leq 1/n_0, \\
 w(y) &= [m + 1 - n(y)][(m + 1)/n(y) - 1]^{-1} \\
 &\quad \times \int_{1-m/n(y)}^{1/n(y)} dx_s \cos[y^{-5}x_s] \cos^{m-1}[y^{-6}/n_0] \cos[y^{-5}\{1 - x_s - (m - 1)/n(y)\}] \\
 &\quad + [n(y) - m][1 - m/n(y)]^{-1} \int_0^{1-m/n(y)} dx_s \cos[y^{-5}x_s] \cos^m[y^{-6}/n_0] \cos[y^{-5}\{1 - x_s - m/n(y)\}]; \\
 m/n_0 \leq y \leq (m + 1)/n_0 &\quad \text{for } m \geq 1.
 \end{aligned} \tag{8}$$

In the limit that  $n_0 \gg 1$ , 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

$$R(n_0) = \frac{\langle 1 - \cos^{n_0 y}[y^{-6}/n_0] \rangle}{\langle 1 - \cos[y^{-5}] \rangle} \sim \frac{\langle 1 - [1 - y^{-12}/2n_0^2]^{n_0 y} \rangle}{\langle 1 - \cos[y^{-5}] \rangle} \sim \frac{\langle 1 - e^{-y^{-11}/2n_0} \rangle}{\langle 1 - \cos[y^{-5}] \rangle} = \frac{0.84}{n_0^{2/11}}, \tag{9}$$

which is the same functional dependence found for the randomly spaced pulses. Note that the form  $\{1 - \cos^{n(y)}[y^{-5}/n(y)]\}$  is identical to that found in theories of the Zeno effect [2–4].

The suppression of decoherence ratio  $R(n_0)$ , obtained from Eqs. (5) 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 due mainly to the fact that the average value of the *square* of the time between pulses in the randomly spaced pulse model is twice that of the equally spaced pulse model [note that the right-hand sides of Eqs. (7) and (9) are in the ratio  $2^{2/11}$ ]. 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.

*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. 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 perturbors. The Weiskopf radius for magnetic decoherence is about 1.0 nm [5], yielding a decoherence rate of  $\approx 10^{10} \text{ s}^{-1}$  at 500 Torr of Xe pressure at 300 °C, and a collision duration  $\tau_c(b_0) \approx 2.5 \text{ ps}$ . 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  $\Delta_s \tau_p$  of order  $2\pi$  and maintain adiabaticity, one can take the detuning  $\delta = 3 \times 10^{13} \text{ s}^{-1}$  and the Rabi frequency  $\Omega \approx 1 \times 10^{14} \text{ s}^{-1}$  on the  $J = 1, m = 0 \rightarrow J = 0$ , excited state transition [6]. The corresponding, power density is  $\approx 1.5 \times 10^{11} \text{ W/cm}^2$ , and the power per pulse is  $\approx 150 \mu\text{J}$  (assuming a  $1 \text{ mm}^2$  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 [2–4]. 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 an experiment of Itano *et al.* [3], 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. Each pulse is said to constitute a measurement, since it is sufficiently long to produce a high likelihood of spontaneous emission whenever the atom is “projected” into the initial state. After each measurement pulse, the off-diagonal density matrix element for the two states of the rf transition is identically equal to zero. In our experiment involving *off-resonant* pulses, the number of Rayleigh photons scattered during each applied pulse is much less than unity. As such, there is no measurement and no quantum Zeno effect, even if suppression of magnetic state decoherence occurs.

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  $n \geq 1$ , 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 occur in a given interval  $T_\pi$ , where  $[x]$  indicates the integer part of  $x$ .

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