# Modification of Atomic Collision Dynamics by Intense Ultrashort Laser Pulses

Theodore Sizer, II,[a] and Michael G. Raymer
*Institute of Optics, University of Rochester, Rochester, New York 14627*
(Received 30 August 1985)

The laser-induced atomic collision process $Na(3S) + Ar + \hbar\omega \rightarrow Na(3P_{1/2}) + Ar$ has been studied by use of an intense, off-resonant laser pulse whose duration is shorter than the collision duration. It has been found that at fixed laser intensity the efficiency for exciting the $Na(3P_{1/2})$ state is higher for these ultrashort pulses than for pulses with duration longer than the collision duration. This effect is caused by a time-dependent modification of the collision potentials by the intense laser field. A theoretical calculation is in qualitative agreement with the results.

PACS numbers: 34.50.Rk

During the past decade there has been a great deal of effort put toward demonstrating that the dynamics of atomic collisions can be modified by the presence of intense laser fields. A large number of theoretical predictions have been reported for a variety of systems,[1-6] but relatively few experiments published to date have shown evidence of such effects.[7-10] The term "modified collision dynamics" means here that the potential energy surfaces which govern the collision dynamics are actually distorted by the ac Stark effect induced by the intense laser field. This results in altered probabilities for the scatterers to end up in certain outgoing channels. This type of strong-field effect should be distinguished from the simple occurrence of an optical collision[11] or a radiative collision,[12] in which the laser merely causes transitions between preexisting and unperturbed collision potentials. The attractiveness of the idea of modified collisions, of course, is that it may present new ways of selectively controlling physical or chemical processes by judicious choice of laser frequency and intensity.

We have taken a novel approach to this problem, as first proposed by Lee and George,[5] by judiciously choosing the duration of the laser pulse to be shorter than the duration of an individual collision. This allows the experimenter to change actively the shape of the potentials during the collision. In principle, if one can open and close reactive channels at appropriate times during the collision, one can strongly influence its outcome. In this paper we report the first experimental observation of the modification of atomic collision dynamics by ultrashort laser pulses.

When two ground-state atoms collide in the presence of an applied laser field of frequency $\omega_L$, a particular interatomic separation $R_c$ exists at which the laser-induced virtual level and the excited-state level intersect, as diagrammed in Fig. 1. Given that the distance of closest approach (impact parameter) for the collision is less than $R_c$, then there are two points in space and time where curve crossings occur. If one of the atoms (the Na atom in this case) ends up in its excited state, it is said that an optical collision,[1,7] or collisional redistribution,[4,6,8,11] has taken place. The time between curve crossings, $T_c$, will be used to define "long" pulses as those laser pulses whose duration is greater than $T_c$, and "short" pulses as those whose duration is shorter than $T_c$. A long pulse, therefore, can interact with both curve crossings whereas a short pulse can interact with only one.

When two levels cross, either two real or a real and a virtual, they avoid one another (in the adiabatic basis) because of the interaction between the two levels. In the case of a virtual level interacting with a real level this interaction is similar to the ac Stark effect, with the smallest separation between the two levels being given by the Rabi frequency $\Omega = dE/h$, where $d$ is the dipole matrix element and $E$ is the laser-electric-field amplitude. This is shown in Fig. 1. The simple Landau-Zener[13] formula can be used to illustrate the principles involved. This formula gives the probability of jumping across the avoided crossing as

$$p = e^{-W}, \tag{1}$$

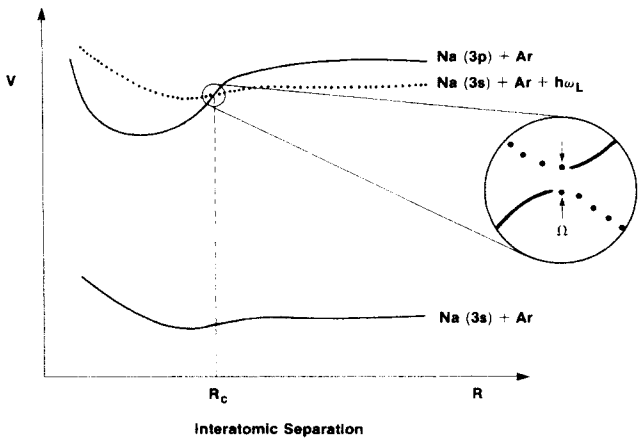


FIG. 1. Simplified schematic of potential energy curves for the collision system Na+Ar. The laser-induced virtual state is shown as a dotted line. The avoided-crossing region is enlarged for clarity.

 

where

$$W = \frac{2\pi\hbar\,\Omega^2}{v|dV_2/dR - dV_1/dR|_{R_c}}, \tag{2}$$

$v$ is the relative velocity, and $V_1$ and $V_2$ are the diabatic (crossing) potentials. The probability of moving from the virtual state to the real excited state in the course of a completed collision is different for the cases of long and short pulses. In the long-pulse case, where the avoided crossing is encountered twice, this probability is given by

$$p = 2e^{-W}(1-e^{-W}), \tag{3}$$

whereas in the short-pulse case the probability is simply

$$p = 1 - e^{-W}, \tag{4}$$

that is, the probability of *not* jumping across the avoided crossing. One can see that at high laser intensities (large values of $W$) there is potentially great advantage in the use of short pulses ($p \to 1$) rather than long pulses ($p \to 0$). This results because at high intensity the collision system adiabatically follows the avoiding potentials. In the case that the long pulse keeps the potentials far apart during the entire collision, the system finds itself back in the initial state following the collision. In the case that the short pulse keeps the potentials far apart during only one of the crossings, the system adiabatically moves onto the excited state and is trapped there.

We have more rigorously treated this problem by solving the two-level Schrödinger equation for straight-line collision trajectories. The equations can be conveniently written in Bloch form as

$$\begin{aligned} \dot{u} &= -[\omega_0(t)-\omega]v, \\ \dot{v} &= [\omega_0(t)-\omega]u + \kappa E(t)w, \\ \dot{w} &= -\kappa E(t)v, \end{aligned} \tag{5}$$

where $u + iv$ is the atomic dipole moment, $w$ is the population inversion, and $\omega_0(t)[V_2(t) - V_1(t)]/\hbar$ is the time-dependent atomic resonance frequency, which was modeled by a simple $C_6/R^6$ interaction.[14] Since the experiment was done in a vapor cell, we had no way of specifying the particular impact parameter nor the interatomic separation between the atoms at the time of the applied-laser-pulse maximum. In order to compare the experiments and the theory, the results of the numerical solution of Eqs. (5) were averaged over possible impact parameters and times between the peak of the laser pulse and the time of closest approach, by use of realistic Gaussian temporal and spatial beam profiles. The result for the expected fluorescence signal from Na($3P_{1/2}$) level is plotted in Fig. 2. Since at a given laser intensity there are more photons incident with use of a longer pulse, in order to compare the probabilities the fluorescence signal was divided by the pulse duration. Although the resulting difference between a long and short pulse is not as dramatic as in Eqs. (3) and (4) because of the effects of averaging, one can still see that there is an advantage to the use of a short pulse over a long pulse. The dramatic intensity dependence of population can be recovered by the assumption of nearly square temporal and spatial beam profiles, as illustrated by the dashed line in Fig. 2.

The system studied experimentally is a vapor of ground-state sodium and argon atoms, excited by picosecond dye-laser pulses tuned 45 $cm^{-1}$ to the low-frequency side of the sodium $D_1$ line at 5896 Å. At this detuning the time between curve crossings is approximately $T_c = 2.5$ psec. At low laser intensities a simple optical (or redistribution) collision takes place during which the collision pair absorbs a photon at a separation $R_c$. This leads to an excited-state sodium atom which radiates at one of the $D$ lines after the collision is completed. On the other hand, at high laser intensities the (adiabatic) potentials are severely distorted in the vicinity of the virtual curve crossing, giving rise to the decrease in excitation efficiency. For long pulses this decrease was observed previously.[7]

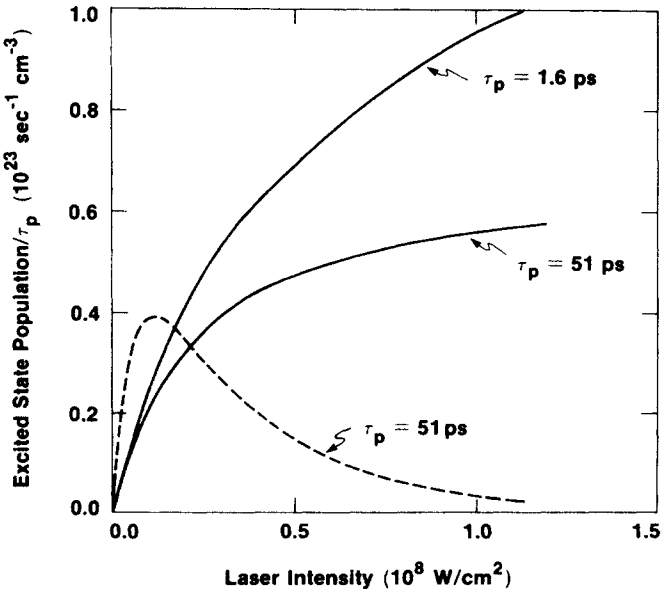


FIG. 2. Solution of the two-level Schrödinger equation for fluorescence intensity divided by pulse duration $\tau_p$, averaged over realistic temporal and spatial profiles, impact parameter, and time between collision and peak of laser pulse. Two laser-pulse durations, 1.6 psec ("short" pulse) and 51 psec ("long" pulse), are shown. The dashed line corresponds to an adiabatic square pulse, with rise and fall times of about 2 psec, and no spatial averaging.

The experimental apparatus is diagrammed in Fig. 3. The output of an amplified picosecond dye-laser system provides pulses of variable duration with energies of 10 $\mu$J at repetition rates up to 1.5 kHz.[15] The beam, after passing through a variable attenuator, was focused into a sodium vapor cell. The diffraction-limited beam at focus had a diameter of 30 $\mu$m. The sodium density was kept low ($5\times10^{12}$ cm$^{-3}$ as measured by the equivalent width) to reduce radiation trapping effects (the optical depth was measured to be 50 $e^{-1}$ depths), while the argon density was kept high (220–250 Torr) to increase the number of collision events. The Na $D_1$-line fluorescence from the cell was imaged onto the slit of a monochromator and the intensity was monitored by a photomultiplier tube (PMT). Care was taken to assure that the signal measured was due to collisional redistribution and not to direct line excitation by amplified spontaneous emission (ASE) generated in the dye amplifier chain at the Na resonance frequency. Experiments done to confirm this included variations of Ar pressure, variation of laser detuning from the resonance line, and blocking the input to the dye amplifier to produce only ASE contributions. Both the laser energy input and the PMT output were measured and tabulated by a computer to find the average output for each laser-energy input. $1.5\times10^4$ shots were accumulated for each pulse duration and the results are plotted together in Fig. 4.

As illustrated in Fig. 4, we have found experimentally a distinct difference in excitation efficiency for different input pulse durations. At a given laser intensity the excitation probability per laser photon is higher for a short (1.6 psec) pulse than for a long (8.5 or 51 psec) pulse, as predicted. However, the observed intensity at which this difference becomes significant is much higher than that predicted by the theory. The discrepancy may be due to the fact that the collision system we used is not a true two-level system as the theory assumes but rather has multiple levels which can be excited.[16] It has been shown theoretically[4] that in the Sr+Ar system the effect of having four levels arising from spatial degeneracy is to diminish the dramatic intensity dependence of fluorescence at high laser intensities predicted in Eq. (3). A discrepancy of similar magnitude has been observed by other researchers for the collision system Tl+Ar, using long pulses.[7]

It might be noticed that a slight turning up of the points on the 51-psec data occurs at the highest powers. This effect, which we believe is statistically significant, is not predicted by the theory. It may be a result of direct, nonadiabatic laser excitation, or a result of the multiple-level nature of the collision. More data on this effect will be presented in a future publication.

In conclusion, we have investigated a laser-induced atomic collision with use of a laser pulse whose duration is shorter than the duration of the collision itself. At high laser field strengths, a pulse-duration dependence of the collision dynamics is predicted theoretically and has been demonstrated experimentally in the collision system Na+Ar. This result offers a dramatic new method for studying atomic collisions by observing the pulse-duration dependence of excitation probability. We conjecture that, for collision systems in which different angular momentum states cross the laser-induced virtual level at different internuclear separations, the polarization of the resulting fluorescence may be pulse-duration dependent. This would arise from essentially the same process studied here: a dependence of outgoing channel on pulse duration.

The authors acknowledge the cooperation of Profes-

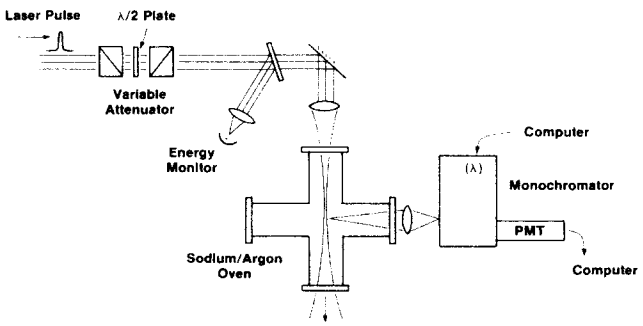


FIG. 3. Experimental setup for detecting Na $D$-line fluorescence resulting from optical collisions between Na and Ar atoms.

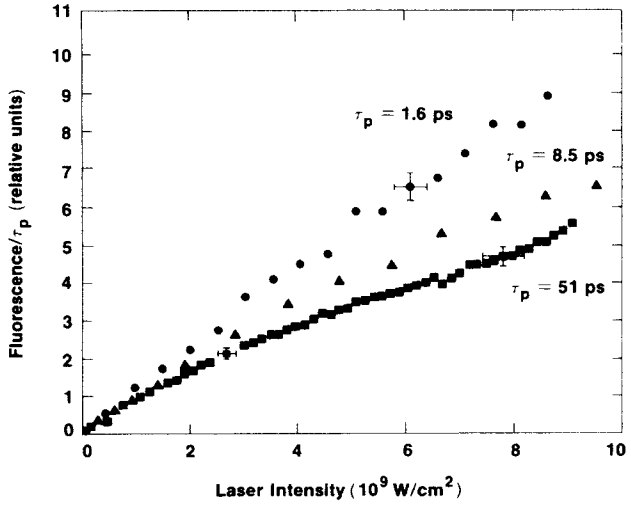


FIG. 4. Experimental results for three different laser pulse widths: 1.6 psec ("short" pulse), and 8.5 and 51 psec ("long" pulse). The dependence on pulse duration indicates a modification of the collision dynamics in the vicinity of the avoided crossing.

sor Gerard Mourou, in whose laboratory these experiments were carried out. The high-speed computer code used for data acquisition was developed with the assistance of Lawrence Forsley. This work was supported by National Science Foundation Physics Grant No. PHY-8215132. This work was also partially supported by the Laser Fusion Feasibility Project at the Laboratory for Laser Energetics, which has the following sponsors: Empire State Electric Energy Research Corporation, General Electric Company, New York State Energy Research and Development Authority, Northeast Utilities Service Company, Southern California Edison Company, The Standard Oil Company (Ohio), and the University of Rochester.

---

[a]Also of the Laboratory for Laser Energetics, University of Rochester. Current address: AT&T Bell Laboratories, Holmdel, N. J. 07733.

[1]V. S. Lisitsa and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. **68**, 479 (1975) [Sov. Phys. JETP **41**, 233 (1975)].

[2]N. M. Kroll and K. M. Watson, Phys. Rev. A **13**, 1018 (1976).

[3]A. M. F. Lau, Phys. Rev. A **13**, 139 (1976).

[4]J. C. Light and A. Szöke, Phys. Rev. A **18**, 1363 (1978).

[5]H. W. Lee and T. F. George, J. Phys. Chem. **83**, 928 (1979).

[6]P. D. Kleiber, J. Cooper, K. Burnett, C. V. Kunasz, and M. G. Raymer, Phys. Rev. A **27**, 291 (1983).

[7]A. M. Bonch-Bruevich, T. A. Vartanyan, and V. V. Khromov, Zh. Eksp. Teor. Fiz. **78**, 583 (1980) [Sov. Phys. JETP **51**, 271 (1980)].

[8]P. D. Kleiber, K. Burnett, and J. Cooper, Phys. Rev. Lett. **47**, 1595 (1981).

[9]W. R. Green, J. Lukasik, J. R. Willison, M. D. Wright, J. F. Young, and S. E. Harris, Phys. Rev. Lett. **42**, 970 (1979); J. Lukasik and S. C. Wallace, Phys. Rev. Lett. **47**, 240 (1981).

[10]P. Pillet, R. Kachru, N. H. Tran, W. W. Smith, and T. F. Gallagher, Phys. Rev. Lett. **50**, 1763 (1983).

[11]J. L. Carlsten, A. Szöke, and M. G. Raymer, Phys. Rev. A **15**, 1029 (1977).

[12]R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, Phys. Rev. A **15**, 1333 (1977).

[13]L. D. Landau, Phys. Z. Sowjetunion **2**, 46 (1932); C. Zener, Proc. Roy. Soc. London A **137**, 696 (1932).

[14]W. R. Hindmarsh, A. D. Petford, and G. Smith, Proc. Roy. Soc. London A **297**, 296 (1967).

[15]I. N. Duling, III, T. Norris, T. Sizer, II, P. Bado, and G. A. Mourou, J. Opt. Soc. Am. **2**, 616 (1985).

[16]A full set of potential curves for this system is not fully known. For an approximate example see M. D. Havey, G. E. Copeland, and W. J. Wang, Phys. Rev. Lett. **50**, 1767 (1985).