Observation of atomic coherence in laser-excited uranium vapor

Donald Scarl
*Polytechnic Institute of New York, Farmingdale, New York, 11735**
and Lawrence Livermore Laboratory, Lawrence, California 94550

Lloyd A. Hackel, Michael A. Johnson, and Michael C. Rushford
Lawrence Livermore Laboratory, Livermore, California 94550
(Received 28 July 1980)

For the pair of ^{238}U levels at 3868 and 26 558 cm^{-1}, the upper-state population was measured as a function of the irradiance of a resonant exciting pulse of fixed duration. The observed damped Rabi flopping showed that the two-level system was partially coherently excited. The results were compared with the prediction of an exact solution of the time-dependent Schrödinger equation for two-level systems excited by a detuned rectangular pulse. The Schrödinger equation solution, after averaging over laser irradiance fluctuations, agrees well with the data.

INTRODUCTION

We have measured the upper-state population as a function of exciting pulse irradiance for a resonantly excited two-level system in atomic uranium vapor. The uranium levels and laser parameters were chosen to maximize the coherence between the upper and lower states in an attempt to observe Rabi flopping. The conditions for partial atomic coherence were optimized by choosing a transition between states of relatively small angular momentum, and by using a narrow-band stable pulsed dye laser, an atomic beam with small Doppler spread, and a uniform laser-beam spatial profile.

The lifetime of the upper state used was long enough that the natural decay could be neglected, making the problem into one of a simple driven two-level system. The uranium atoms are in the lower state at the beginning of the laser pulse, are excited into the upper state and back into the lower state, more or less, during the laser pulse, and remain, after the pulse is over, in a state determined by the height and duration of the laser pulse. A later ionizing pulse allows the upper-state population to be measured. The upper-state population was measured as a function of the height of the exciting pulse. The width of the exciting pulse was held constant.

The measured upper-state population was compared with the predictions of an exact solution of the time-dependent Schrödinger equation for a set of two-state systems driven by a detuned laser.[1-4] Although the average laser detuning was zero, the spectral width of the laser was taken into account by assuming a mean-square detuning in the final expression for the upper-state population. The pulse-to-pulse fluctuation in laser irradiance was averaged over, leading to a Gaussian damping of the Rabi oscillation.

Although Rabi flopping in an atomic two-state system has been observed before,[5] this is the first time that the upper-state population has been measured as a function of the height rather than the width of the exciting pulse. When the exciting pulse has a spectral width that is comparable with the Rabi frequency, the shape of the population versus irradiance curve is different from that of the population versus pulse length curve.

This is also the first time that coherent atomic effects have been seen in the complex uranium atom, that flopping has been seen in a system excited by a short-pulse dye laser, and that ion detection rather than fluorescence has been used to measure the population of a coherently excited upper state.

From observation of coherent excitation of the upper state the matrix element connecting the two states can be determined. For the uranium states at 3868.486 and 26 557.936 cm^{-1}, the dipole matrix element for a $m = 0$ to $m = 0$ transition was measured to be $(4.1 \pm 0.6) \times 10^{-12}$ m.

URANIUM ENERGY LEVELS

The relevant levels in ^{238}U are shown in Fig. 1. The lower level of the two-level system was the $j = 3$ level at 3868 cm^{-1}. The upper level was the $j = 2$ level at 26 558 cm^{-1}. Although at the temperature of the oven, only 3% of the atoms in the beam were in the 3868 cm^{-1} state, the great sensitivity provided by ion detection allowed this state to be used as the initially populated level of a two-level system. The subsequent population of the 26 558 cm^{-1} level was measured as a function of the irradiance of the 440.61-nm laser pulse connecting the two levels.

CALCULATED UPPER STATE POPULATION

Two-level system

The driven two-level system is one of the few quantum-mechanical systems for which the time-

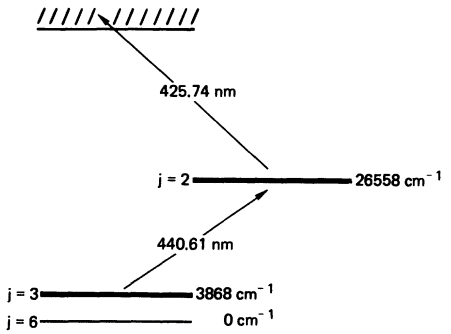

FIG. 1. The ^{238}U levels used in the experiment. For linearly polarized light, the five $\Delta m = 0$ transitions from the $j = 3$ to the $j = 2$ states can be analyzed as five similar independent two-level systems.

dependent Schrödinger equation can be solved exactly. The Schrödinger equation is

$$H|\psi\rangle = i\hbar \frac{\partial}{\partial t}|\psi\rangle , \tag{1}$$

with matrix elements

$$\langle 1|H|1\rangle = \hbar\omega_1 , \quad \langle 2|H|2\rangle = \hbar\omega_2 , \tag{2}$$

$$\langle 2|H|1\rangle e^{-i\Delta t} = \frac{eEz}{2} = \frac{\hbar\Omega_0}{2} , \tag{3}$$

where E is the amplitude of the driving field, ez is the dipole matrix element between the two states, and Ω_0 is the resonant Rabi frequency of the interaction. The laser frequency is ω and its offset from the resonance frequency is $\Delta = (\omega_2 - \omega_1) - \omega$.

The state of the system at any time is

$$|\psi\rangle = c_1|1\rangle + c_2 e^{i\Delta t}|2\rangle . \tag{4}$$

For an atom in the lower state at $t = 0$, the probability of being in the upper state at later times is[1-4]

$$P_2 = |c_2|^2 = \frac{1}{2}\left(\frac{\Omega_0}{\Omega}\right)^2 (1 - \cos\Omega t) , \tag{5}$$

where $\Omega = \Omega_0^2 + \Delta^2$.

Laser fluctuations

The probability calculation leading to Eq. (5) assumed that the detuning Δ and the resonant Rabi frequency Ω_0 were constants. In the experiment, however, the detuning varied about a mean value of zero because of the frequency spread of the laser beam. In evaluating the theoretical upper-state probabilities this mean value was used for Δ^2. (Δ itself does not appear in the probability expression.)

The spread in Ω_0 caused by the spatial and pulse-to-pulse variation of the irradiance of the laser beam was taken into account by averaging P_2 using a Gaussian distribution of laser irradiance H with a width σ_H proportional to $\langle H\rangle$. As described in the Appendix, this average, for values of $s = \sigma_H/\langle H\rangle < 1$ yields

$$\langle P_2\rangle = \frac{1}{2}\left(\frac{\Omega_r}{\Omega}\right)^2 \left[1 - \exp\left(\frac{-(\Omega_r t)^2}{2\sigma^2}\right)\cos\Omega t\right] , \tag{6}$$

where

$$\Omega^2 = \Omega_r^2 + \Delta^2 , \quad \sigma = 2\Omega/s\Omega_r . \tag{7}$$

Ω_r is the root-mean-square value of Ω_0. The average upper-state probability shows coherent excitation (Rabi flopping or optical nutation) for small values of $\Omega_r t$ and incoherent excitation (saturation) for large values of $\Omega_r t$.

Coherence between the upper and lower atomic states can also be lost by upper-state decay through spontaneous emission, and upper-state decay and dephasing caused by atomic collisions.[3,4,6] In addition, even lasers with well regulated amplitudes exhibit phase diffusion[7,8] and ultimately, the quantum fluctuations that accompany a coherent state of the radiation field.[9-11] In the present experiment, the largest contributor to loss of atomic coherence is the pulse-to-pulse fluctuation of the dye-laser irradiance. Compared with this fluctuation, the upper-state lifetime is long, the atoms in the beam suffer few collisions, and the quantum-field fluctuations are negligible.

Level degeneracies

For a $j = 3$ to $j = 2$ transition driven by linearly polarized light, five different $\Delta m = 0$ transitions are possible, with the matrix elements[12]

$$\begin{aligned} &z_0 , \quad \text{for } m = 0 \to m = 0 , \\ &z_1 = \sqrt{8/9}\, z_0 , \quad \text{for } m = \pm 1 \to m = \pm 1 , \\ &z_2 = \sqrt{5/9}\, z_0 , \quad \text{for } m = \pm 2 \to m = \pm 2 . \end{aligned} \tag{8}$$

Since the states of different m are uncoupled, the upper-state probabilities add, leading to a total upper-state probability of

$$P_2 = \tfrac{1}{2} \sum_{i=0,1,2} b_i \left(\frac{\Omega_{ri}}{\Omega_i}\right)^2 \left[1 - \exp\left(\frac{-(\Omega_{ri} t)^2}{2\sigma_i^2}\right)\cos\Omega_i t\right] , \tag{9}$$

with

$$\Omega_{ri} = (\langle \Omega_{0i}^2\rangle)^{1/2} = eEz_i , \quad \Omega_i^2 = \Omega_{ri}^2 + \Delta^2 \tag{10}$$

and

$$b_0 = \tfrac{1}{7} , \quad b_1 = b_2 = \tfrac{2}{7} . \tag{11}$$

The weight functions b_i take into account the single state with $m = 0$, two states each for $m = \pm 1$ and $m = \pm 2$, and a total of seven degenerate m states

for the $j=3$ lower level. For linearly polarized excitation, the maximum probability for being in the upper state of a $j=3$ to $j=2$ transition is $\frac{5}{7}$.

If the exciting electric field pulse is approximated as rectangular with a height E and a width t, P_2 is the probability that the atom will be in the upper state at the end of the exciting pulse. This probability, Eq. (9), is plotted as a function of E in Fig. 3(b). The effective pulse height, width, and frequency spread are calculated in the Appendix.

EXPERIMENT

A uranium rhenide source was heated in a tungsten oven to 1750 K and the resulting uranium vapor passed through two 1-mm wide 10-mm long slits separated by 20 mm (Fig. 2). The two laser beams intersected the atomic beam 25 mm from the second slit at which point the uranium atomic density was approximately 10^9 atoms per cm^3. The bisector of the 5° angle between the two laser beams was perpendicular to the atomic beam. The residual atomic Doppler width was $\sigma_D = 56$ MHz.

The exciting 440.61-nm laser was a pulsed dye laser[13] pumped by a commercial Molectron nitrogen laser. The dye was C440 mixed to a concentration of 1×10^{-4} molar in ethanol. The nitrogen laser was operated at a repetition rate of 19 Hz and had a peak power of 300 MW at 337 nm in a pulse whose full width at half maximum (FWHM) was approximately 10 ns. The electric vector of both the pumping laser beam and the dye-laser beam were vertical. The dye-laser cavity consisted of a fully reflecting plane mirror, a 10 mm-long dye cell, a 3 mm-thick solid etalon, a 10-mm prism used as a beam expander, and a 2100 lines per mm grating. The spacing between mirror and grating was 70 mm, leading to a free spectral range of 2 GHz. The laser was tuned by tilting the grating and etalon. Preliminary setting of the laser to the uranium line was done by comparing the laser wavelength with the appropriate line from an electroless discharge uranium lamp

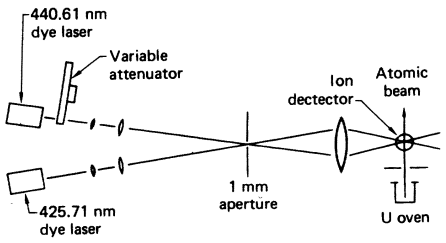

FIG. 2. The experimental arrangement. The ion current was measured as a function of the irradiance of the 440.61-nm laser.

using a 1.5-m Jobin Yvon monocrometer with a silicon vidicon detector. Final setting of the laser frequency to the center of the atomic line was done by maximizing the number of ions detected.

The laser pulse width was measured to be 6.0 ns (full width at half maximum) using a 1 ns-risetime oscilloscope and 1 ns-risetime vacuum photodiode. The effective pulse width is calculated in the Appendix. The width of the spectral distribution of the laser was measured to be 180 MHz FWHM using a Fabry Perot interferometer with a free spectral range of 300 MHz and a finesse of 20. The effective mean-square frequency deviation is calculated in the Appendix and was used in evaluating Eq. (9).

The laser beam passed through a circular variable attenuator with a density range from 0 to 2.0. This attenuator rotated at 0.1 rps, providing a factor of 100 change in the exciting beam irradiance every 10 s. A telescope increased the beam diameter to 10 mm and brought the beam to a gradual focus so that the beam diameter at a plane 3 m from the telescope was 6 mm. A 1 mm-diameter aperture in this plane selected only the relatively uniform central part of the roughly Gaussian spatial profile. The irradiance was uniform within ±20% over the 1-mm aperture. The 1-mm aperture was focused with a magnification of 0.44 into the vacuum chamber containing the atomic beam.

The 425.74-nm ionizing laser was a modified Molectron dye laser, pumped by a second Molectron N_2 laser. The spectral width of the light from this laser was approximately 3 GHz. Its irradiance of 10^9 W/m^2 in a 5-ns pulse was able to saturate the transition from the uranium 26 558-cm^{-1} level to an autoionizing level in the continuum so that the number of ions produced was independent of small power fluctuations of the ionizing laser. The number of detected ions was also independent of the delay between the exciting and ionizing lasers for delays between about 5 and 30 ns.

Ions produced by the two lasers were accelerated by a 15-V potential across two plates separated by 10 mm and centered on the uranium-beam–laser-beam intersection point. After passing through a fine mesh screen covering a 10 mm-diameter aperture in the upper plate, they were detected by a Galileo Channeltron electron multiplier which produced a pulse of about 10^6 electrons for each incident ion. The Channeltron output pulse was integrated over the 10 μs that it took all of the ions produced in one laser pulse to reach the detector, amplified, and averaged over approximately three laser pulses in a boxcar averager. The signal from the boxcar averager was further

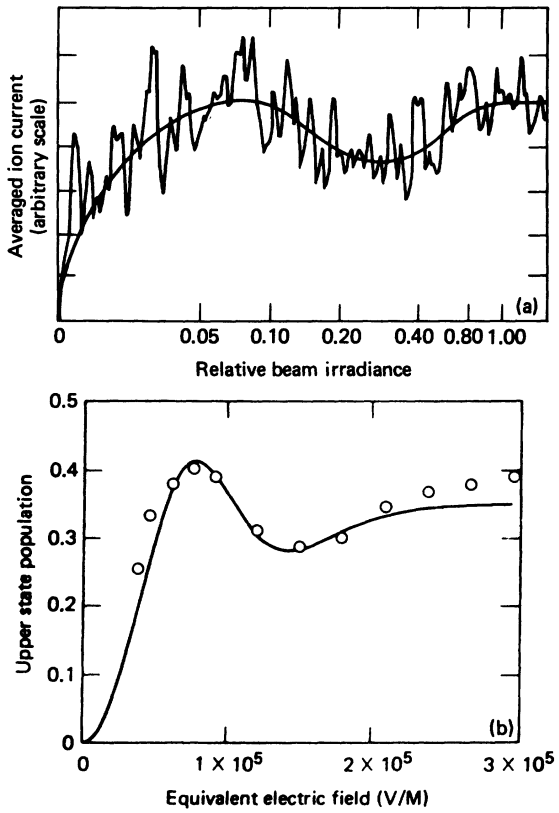

FIG. 3. (a) The measured ion current as a function of irradiance. The rapidly varying curve is the data from the signal averager. The smooth curve is an average of the data curve and was used to produce the points of (b). (b) Upper-state population vs laser electric field. The points are the data taken from (a). The solid curve is a plot of Eq. (9), which is a solution of the Schrödinger equation averaged over the fluctuating laser irradiance.

averaged by a Nicolet signal averager whose sweep period was synchronized with the rotating variable attenuator. For each signal averager run about 100 sweeps were averaged over a 3 min period, during which time the variable density filter rotated about 30 times and the laser was pulsed approximately 3500 times.

At the beginning of each sweep an opaque region of the attenuator wheel allowed determination of the zero level of the ion signal. At the end of each sweep a clear area on the attenuator wheel allowed centering of the laser frequency on the atomic transition frequency by maximizing the number of ions detected during the several seconds that the exciting laser irradiance was unchanging.

RESULTS

One of several similar runs is shown in Fig. 3(a). The horizontal scale is linear in time and corresponds to one rotation of the variable neutral density filter in front of the exciting laser. The density of the filter is roughly linear with rotation angle, except for a small area near the beginning of each cycle for which the density is infinite and for the last 90° of rotation over which the density is constant and approximately zero. Over the linear region the laser irradiance increases exponentially. The actual irradiance was measured with a linear photodiode and used to calibrate the horizontal scale in Fig. 3(a). The vertical scale is a relative scale proportional to the number of ions detected. The plotted curve is taken directly from the signal averager.

Figure 3(b) is a replotting of the smoothed data curve on a scale linear in the laser electric field strength. [The points of Fig. 3(b) were taken from the smoothed curve of Fig. 3(a).] On the same scale the theoretical upper state population, Eq. (9), is plotted as a solid line. The constants that lead to the satisfactory fit shown are

$$z_0 = (4.1 \pm 0.6) \times 10^{-12}\ \text{m}\,, \qquad s = 0.7 \pm 0.1\,. \tag{12}$$

CONCLUSIONS

We have shown that atoms whose lifetime is longer than the Rabi time can be coherently excited by pulsed dye lasers and that the coherence is measurable even in the presence of considerable level degeneracy and laser irradiance fluctuations. The coherent Rabi flopping of the upper-state population can be seen over a small range of laser irradiance. For large values of irradiance the coherent flopping goes smoothly over into incoherent saturation.

ACKNOWLEDGMENTS

We thank Bruce Shore, Richard Solarz, James Holtz, and James Davis for useful discussions and encouragement.

APPENDIX

Equivalent rectangular laser pulse

The time dependence of the laser pulse, which was roughly Gaussian with peak height P_g and width σ_g, was approximated by a rectangular pulse of height P and width t in order to be able to solve the time dependent Schrödinger equation, Eq. (1), exactly. P and t were determined from the measured P_g and σ_g by requiring that the zeroth, first, and second moments of the rectangular pulse be equal to those of the Gaussian pulse. If both first moments are set equal to zero, the equality of zeroth moments and of second moments lead to

$$t = 2\sqrt{3}\,\sigma_g\,, \tag{A1}$$

$$P=\sqrt{\pi/6}\,P_g\,. \tag{A2}$$

The measured width of the Gaussian shaped laser pulse was 2.55 ns, leading to a pulse width of $\sigma_g = 2.13$ ns after deconvolution from the 1.5-ns system impulse response. From Eq. (A1), this leads to $t = 7.38$ ns for the pulse width of the equivalent rectangular pulse.

The height of the Gaussian laser pulse, with minimum attenuation in the beam, was 20 W, which after deconvolution gave an actual peak power of 24 W. This height P_g led to a height for the equivalent rectangular pulse of $P = 17.4$ W. Since the laser beam in the interaction region had a radius of 2.22×10^{-4} m, the equivalent irradiance was 1.12×10^{8} W/m^2 and the equivalent electric field was 2.91×10^{5} V/m. The field seen by the atoms went from zero to this value for each rotation of the variable attenuator.

Effective detuning

A Gaussian power pulse with a width $\sigma_g = 2.13$ ns has a Gaussian power spectrum with a transform limited spectral width of $\sigma_\omega = 2.36\times10^{8}$ rad/s. If the width of the power spectrum of the laser pulse had this value, the atoms would notice no detuning and Δ would be zero. However, the measured width of the power spectrum was $\sigma_\omega = 4.87\times10^{8}$ rad/s. The effective detuning was taken to be the difference between the measured spectral width and the transform limited spectral width, leading to $\Delta = 2.51\times10^{8}$ rad/s.

Average over laser pulse heights

With no attenuation in the beam, the laser pulse-to-pulse irradiance fluctuations were assumed to have a Gaussian probability distribution

$$P(H)=\frac{1}{\sqrt{2\pi}\,\sigma_H}\exp\left(\frac{-(H-\langle H\rangle)^2}{2\sigma_H^2}\right), \tag{A3}$$

where $\langle H\rangle$ and σ_H are the average value and standard deviation of the laser irradiance. As the beam is attenuated the ratio $s=\sigma_H/\langle H\rangle$ remains constant. With

$$\frac{\Omega_0^2}{H}=\frac{\Omega_r^2}{\langle H\rangle}=\frac{\sigma_r}{\sigma_H}=\frac{2}{\epsilon_0 c}\left(\frac{ez}{\hbar}\right)^2,\quad \frac{\sigma_H}{\langle H\rangle}=\frac{\sigma_r}{\Omega_r^2}=s\,, \tag{A4}$$

the upper-state probability averaged over laser irradiance fluctuations is

$$\langle P_2\rangle=\frac{1}{2\sqrt{2\pi}\,\sigma_r}\int_0^\infty \exp\left(\frac{-(\Omega_0^2-\Omega_r^2)^2}{2\sigma_r^2}\right)\left(\frac{\Omega_0}{\Omega}\right)^2 \times(1-\cos\Omega t)\,d\Omega_0^2\,. \tag{A5}$$

Although this can be integrated numerically, it can be simplified considerably when $s<1$ since the narrow Gaussian restricts Ω_0 to a small range. With the substitutions

$$x^2=\Omega_0^2+\Delta^2\,,\quad x_r^2=\Omega_r^2+\Delta^2\,, \tag{A6}$$

$$\langle P_2\rangle=\frac{1}{\sqrt{2\pi}\,\sigma_r}\int_\Delta^\infty \exp\left(-\frac{(x-x_r)^2(x+x_r)^2}{2\sigma_r^2}\right) \times\left(\frac{x^2-\Delta^2}{x}\right)(1-\cos xt)\,dx\,. \tag{A7}$$

If x is set equal to x_r everywhere except in the first term in the exponential and in the cosine, the integral reduces to

$$\langle P_2\rangle=\frac{1}{2}\,\frac{\Omega_r^2}{\Omega_r^2+\Delta^2}\left[1-\exp\left(-\frac{(\Omega_r t)^2}{2\sigma^2}\right)\cos[(\Omega_r^2+\Delta^2)^{1/2}t]\right], \tag{A8}$$

where

$$\sigma=\frac{2(\Omega_r^2+\Delta^2)^{1/2}}{s\Omega_r}\,. \tag{A9}$$

This is the simple Rabi flopping expression with exponential damping of the cosine term. The exponential damping expresses the partial coherence of the atomic states $|1\rangle$ and $|2\rangle$ (just as a similar term in a two-beam interference experiment with partially coherent light expresses the partial coherence of the two beams).

*Present address.

[1] J. H. Eberly, B. W. Shore, Z. Bialynicka-Birula, and I. Bialynicki-Birula, Phys. Rev. A 16, 2038 (1977); 16, 2048 (1977).

[2] Bruce W. Shore, Phys. Rev. A 17, 1739 (1978).

[3] J. R. Ackerhalt, J. H. Eberly, and B. W. Shore, Phys. Rev. A 19, 248 (1979).

[4] P. L. Knight and P. W. Milonni, Phys. Rep. 66, 21 (1980).

[5] H. M. Gibbs, Phys. Rev. A 8, 446 (1973).

[6] J. H. Eberly, Phys. Rev. Lett. 37, 1387 (1976).

[7] Paul Avan and Claude Cohen-Tannoudji, J. Phys. B 10, 155 (1977).

[8] G. S. Agarwal, Phys. Rev. A 18, 1490 (1978).

[9] F. W. Cummings, Phys. Rev. 140, A1051 (1965).

[10] P. Meystre, A. Quattropani, and H. P. Baltes, Phys. Lett. 49A, 85 (1974).

[11] J. H. Eberly, N. B. Narozhny, and J. J. Sanchez-Mondragon, Phys. Rev. Lett. 44, 1323 (1980).

[12] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1935).

[13] D. C. Hanna, P. A. Karkkainen, and R. Wyatt, Opt. Quantum Electron. 7, 115 (1975).