RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 27, NUMBER 3 MARCH 1983

Spin polarization by selective laser-induced interference

G. Alber
Institute for Theoretical Physics, University of Innsbruck, 6020 Innsbruck, Austria

P. Zoller*
Joint Institute for Laboratory Astrophysics, National Bureau of Standards and University of Colorado, Boulder, Colorado 80309

(Received 3 December 1982)

States behaving like autoionizing states can be selectively induced by laser radiation into one of the continuum spin channels in the photoionization of polarized excited alkaki atoms. As a result of destructive or constructive interference between the direct ionization channel and those introduced by the dressing laser radiation, the cross section of this specific spin component is completely suppressed or enhanced, while leaving the other spin channel unaffected. The q parameter which determines the line shape of the Fano-type resonance can be resonantly tuned as a function of the dressing laser frequency.

Laser radiation can be used to enhance or suppress reaction channels by inducing autoionizinglike resonances to a given energy in the continuum[1–6]; in these cases the laser does not contribute directly to the dynamics of the multichannel process, but rather dresses the final states of the system. Constructive and destructive interference between the original reaction paths and those induced by the dressing radiation leads to enhanced or suppressed cross sections to the various final states of the reaction. Recent experiments have demonstrated enhanced harmonic generation[5] and rotation of the light polarization of a probe beam[6] in the neighborhood of these induced resonances.

In the present Rapid Communication we propose the production of spin-polarized electrons in resonant multiphoton ionization by *selectively* inducing autoionizinglike resonances in one spin channel, while leaving the cross section for the other components unaffected. At the minimum of these Fano-type resonances[7] this particular spin channel is suppressed by destructive interference resulting in 100% spin-polarized electrons. At the same time the line-shape parameter of the Fano resonance can be resonantly tuned as a function of the dressing laser frequency.

We consider photoionization from an excited state of an alkali atom by right circularly polarized light.[8] To be more specific, we discuss in the following the scheme given in Fig. 1. According to the selection rules in multiphoton ionization summarized in Ref. 9, two-photon excitation form the $^2S_{1/2,m}$ ($m=\pm\frac{1}{2}$) states with right circularly polarized light populates the $^2D_{3/2,3/2}$ and $^2D_{5/2,m}$ ($m=\frac{3}{2}$ and $\frac{5}{2}$) Zeeman states. We assume the hyperfine structure of the ground state to be much smaller than the fine-structure splitting of the ^2D state. In addition we assume that the laser does not resolve the hyperfine structure of the excited-state doublets.

A laser will only resonantly populate the $^2D_{3/2,3/2}$ state, while resonant excitation of the $^2D_{5/2}$ level produces atoms both in the $m=\frac{3}{2}$ and $\frac{5}{2}$ states. Note that according to angular momentum recoupling, $^2D_{5/2,5/2}$ is a pure spin-up state while $^2D_{5/2,3/2}$ and $^2D_{3/2,3/2}$ are a mixture of spin states. Accordingly, single-photon ionization of the $^2D_{5/2}$ and $^2D_{3/2}$ levels with a right circularly polarized probe laser will produce spin-up polarized electrons from the $m=\frac{5}{2}$ state in the $^2F_{7/2,7/2}$ continuum, while ionization from $m=\frac{3}{2}$ will result in a mixture of spin states in the $^2F_{5/2,5/2}$ and $^2F_{7/2,5/2}$ continua.[9] To suppress or enhance the ionization signal in the spin-up or spin-down channel, we induce—with the help of an intense dressing laser—a second ionization channel to the continuum: We excite with an intense laser of frequency ω_d one of the higher-lying states

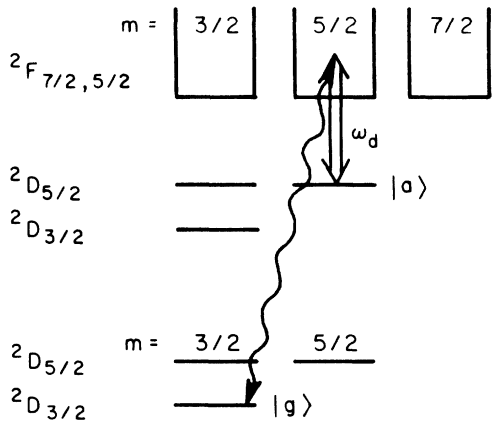

FIG. 1. Excitation scheme.

($|^2Djm\rangle \equiv |ajm\rangle$, for example) by an induced Raman process where a probe laser photon ω_p is absorbed and a photon ω_d is emitted (or vice versa when excited ^2D is populated). Ionization from this Raman excited state by the strong (dressing) laser provides an second ionization channel which coherently interferes with the direct ionization process and is responsible for Fano-type resonances[7] in the ionization continua. The frequency ω_d is chosen so that direct ionization from the two-proton excited ^2D ($\equiv |g\rangle$) state is negligible. The ionization rate of an excited state $|g\rangle$ into the spin-up ($\mu=+\frac{1}{2}$) and spin-down channels ($\mu=-\frac{1}{2}$) is in lowest-order perturbation theory of the probe laser[10] given by

$$W_\mu=\frac{2\pi}{\hbar}\sum_{lm}\left|\langle E\mu lm|D_p^+|g\rangle+\sum_{jm_j}\frac{\langle E\mu lm|D_d^+|ajm\rangle}{\Delta_{jm}-i(\hbar/2)\Gamma_{jm}}\langle ajm|R|g\rangle\right|^2 , \tag{1}$$

where

$$\langle ajm|R|g\rangle=\left\langle ajm\left|D_d^-\frac{1}{E_g+\hbar\omega_p-H_A+i\epsilon}D_p^+\right|g\right\rangle+\left\langle ajm\left|D_p^+\frac{1}{E_g-\hbar\omega_d-H_A}D_d^-\right|g\right\rangle \tag{2}$$

is the Raman transition matrix element. Note that for low-lying atomic states $|g\rangle$ the contribution from the second term in Eq. (2) will usually be small.

The energy normalized continuum wave functions are denoted by $|E\mu lm\rangle$ neglecting spin-orbit effects in the continuum.[9] $D_p^{(\pm)}$ and $D_d^{(\pm)}$ are the dipole interaction operators describing absorption (emission) of a photon with frequency ω_p and ω_d, respectively. The first term in Eq. (1) describes the direct ionization process while the second corresponds to a Raman excitation followed by ionization. H_A is the atomic Hamiltonian.

The total induced linewidth of levels $|ajm\rangle$ due to ionization by the dressing laser is

$$\Gamma_{jm_j}=\frac{2\pi}{\hbar}\sum_{lm\mu}|\langle E\mu lm|D_d^+|ajm_j\rangle|^2 . \tag{3}$$

Spontaneous decay and other incoherent contributions have been ignored in Eq. (1). $\Delta_{jm}=E_g+\hbar\omega_p-E_{ajm}-\hbar\omega_d$ is the detuning from the Raman resonance including the dynamic Stark shifts in the definitions of the atomic energies. Equation (1) must be summed over the populations of the two-photon excited states.

In general, the interference between the ionization paths in Eq. (1) is incomplete and will lead to Fano-type resonance superimposed onto a background in both spin channels. This is due to the many continua available as final states in the ionization process [the sum over the angular momentum quantum numbers in Eqs. (1) and (3)] as well as the intermediate Raman states. For specific configurations (Fig. 1), however, a complete destructive interference which selectively suppresses one of the spin channels, while leaving the other one unaffected, is possible. Consider, for example, ionization from a $^2D_{3/2,3/2}$ ($|g\rangle$) state. Choosing a linearly polarized dressing laser, a Raman process starting from $|g\rangle$ will only excite the $^2D_{5/2,5/2}$ ($|a\rangle$) state which is a pure spin-up state. The intense laser will ionize this state selectively into the spin-up $|E\mu=\frac{1}{2},l=3,m=2\rangle$ continuum, which again is the only spin-up continuum populated from level $|g\rangle$. The ionization signal in the spin-up channel exhibits, therefore, a Fano profile,

$$W_+=\gamma_+\frac{(\epsilon+q)^2}{\epsilon^2+1} , \tag{4}$$

where γ_+ is the ionization rate of $|g\rangle$ in the presence of the probe laser alone; $\epsilon=\Delta/(\frac{1}{2}\hbar\Gamma)$ is the scaled detuning from the Raman resonance and

$$q=-\mathrm{Re}(R_{ag})/\mathrm{Im}(R_{ag}) \tag{5}$$

is defined as the quotient of the real and imaginary parts of the Raman matrix element (2). At the same time, ionization from $|g\rangle$ resulting in spin-down electrons is unaffected by this laser-induced interference: We find $W_-=\gamma_-$ with γ_- the unperturbed ionization rate of $|g\rangle$. The spin polarization defined by

$$P=(W_+-W_-)/(W_++W_-)$$

shows, as a function of ϵ, complete spin-down polarization for $\epsilon=-q$ and mainly spin-up polarized electrons for $\epsilon\approx 0$ and $q\gg 1$ due to destructive and constructive interference (Fig. 2).

The parameter q determines the shape of the resonance. It is independent of the light intensity, but shows a resonance structure as a function of the laser frequency ω_d: The counter-rotating Raman term [i.e., the second term in Eq. (2)], which has been ig-

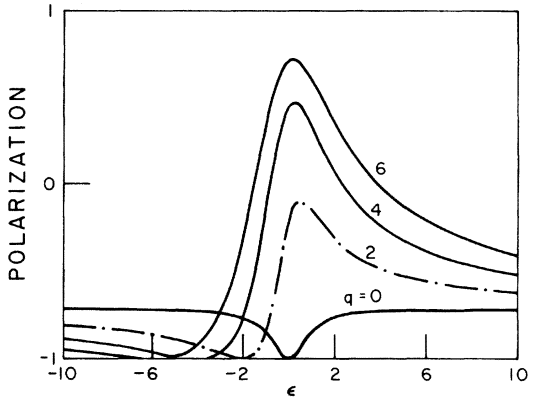

FIG. 2. Spin polarization P as a function of the scaled detuning ϵ for $\gamma_+/\gamma_-=\frac{1}{6}$ and $q=0$, 2, 4, and 6.

nored in previous work involving autoionizinglike states, has intermediate resonances whenever $\hbar\omega_d$ matches the energy difference between $|g\rangle$ and a lower-lying state coupled to it by a dipole transition. Around these resonances q shows a dispersive behavior superimposed on a (usually) slowly varying background originating from the nonresonant terms (Fig. 3). The parameter q changes sign on these intermediate resonances (where it diverges in our approximation). As the background from the nonresonant terms adds constructively on one and destructively at the other side of the resonance, q can be expected to have a zero at a certain frequency ω_d. At this frequency the real part of the Raman matrix element goes through zero and the Fano profile has the form of a window resonance. Besides the possibility of controlling the line shape of the resonance with the frequency of the dressing laser, a measurement of q provides information on the sign and magnitude of the Raman transition matrix element. Figure 3 illustrates this behavior for a $5^2D_{3/2}$-$15^2D_{5/2}$ Raman transition in cesium. We note that at the same time the Raman transition has an intermediate resonance, the Stark shift of $|a\rangle$ will also exhibit a dispersive behavior.

If the counter-rotating Raman term is neglected in Eq. (2), the effect of the dressing laser can be viewed as inducing an autoionizinglike resonance in the final state of the photoionization process. In this picture, the counter-rotating term may be described as an initial state interaction, i.e., a laser-induced dressing of the state $|g\rangle$.

The physical mechanism behind our spin-polarization scheme is the spin-orbit splitting of the ^2D level, which serves as the ground state for the ionization process, combined with a selective laser-induced interference. Andryushin and Fedorov[3] proposed to produce spin-polarized electrons by inducing autoionizinglike states from an excited $^2S_{1/2}$ alkali state in the $^2P_{1/2}$ and $^2P_{3/2}$ continua by right circularly polarized light. This induces resonances in both the $m=\frac{1}{2}$ and $\frac{3}{2}$ continua. Due to the different spin-orbit dependence of the bound-free matrix elements, and the different vector polarizabilities of the $^2S_{\pm 1/2}$ states (both of which have been ignored in our calculation), the minima of the Fano profiles will be slightly shifted relative to each other, which, in analogy to the Fano effect,[11] will result in almost complete spin polarization for ionization from the ground state in this energy range. In our scheme, an autoionizinglike resonance is introduced selectively in the spin-up continuum; the cross section of the spin-down component does not have a minimum near these energies. Spin-orbit splitting of the excited states of an alkali atom is also responsible for the spin polarization of electrons in nonresonant multiphoton ionization with circularly polarized light as suggested by Lambropoulos.[12] There, the ionization

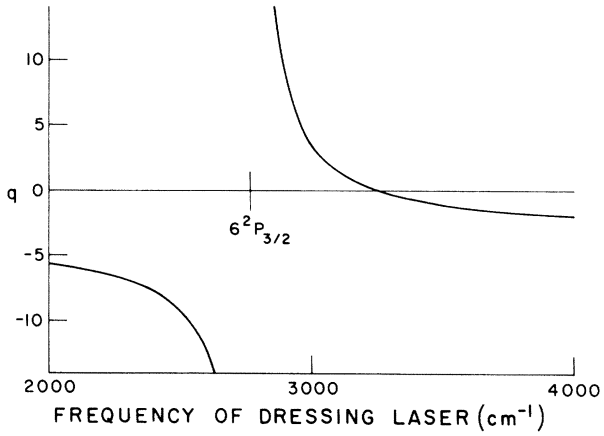

FIG. 3. The q parameter as a function of the dressing laser frequency ω_d (cm^{-1}) and for the $5^2D_{3/2,3/2}$ to $15^2D_{5/2,5/2}$ transition in cesium, estimated by a truncated summation over the intermediate atomic states in Eq. (2) using quantum-defect wave functions. q shows as resonance when the photon energy of the dressing laser matches the energy difference form the $5^2D_{3/2}$ state to the lower-lying $6^2P_{3/2}$ level, which is one of the intermediate states contributing to the second Raman term in Eq. (2).

channel from the $^2S_{-1/2}$ ground state, which gives a mixture of spin states, can be suppressed by destructive interference when the laser is tuned between the fine-structure levels.[9,12,13] Contrary to the process discussed above, in this case the interference responsible for spin polarization is part of the excitation dynamics of the process itself and not induced by an external light source.

As the present spin polarization effect is based on a laser-induced interference, any incoherent mechanism such as spatial or temporal intensity variations of the dressing laser, admixture from other ionization channels, spontaneous decay, and partially resolved hyperfine structure of the excited state will tend to destroy the effect.

In the present Communication we have demonstrated in the case of spin polarization that laser radiation can be used to selectively enhance or suppress certain reaction channels at a given energy. This suggests the use of selectively induced autoionizinglike resonances to control the branching ratios between various final states in a multichannel reaction, either to increase the efficiency in the production of a desired final state or to improve the signal-to-background ratio when measuring small cross sections in the background of a dominating process.

ACKNOWLEDGMENTS

P.Z. thanks Dr. K. Burnett, Dr. J. Copper, Dr. P. L. Knight, and Dr. E. Matthias for helpful discussions. G.A. acknowledges financial support by the Austrian National Bank.

*JILA visitor; permanent address: Institute for Theoretical Physics, University of Innsbruck, 6020 Innsbruck, Austria.

[1]L. Armstrong, Jr., B. L. Beers, and S. Feneuille, Phys. Rev. A 12, 1903 (1975); B. L. Beers and L. Armstrong, Jr., *ibid.* 12, 2447 (1975).

[2]Yu. I. Heller and A. K. Popov, Opt. Commun. 18, 449 (1976).

[3]A. I. Andryushin and M. V. Fedorov, Zh. Eksp. Teor. Fiz. 75, 2037 (1978) [Sov. Phys. JETP 48, 228 (1978)].

[4]Yu. I. Geller and A. K. Popov, Zh. Eksp. Teor. Fiz. 78, 506 (1980) [Sov. Phys. JETP 51, 255 (1980)].

[5]L. I. Pavlov, S. S. Dimov, D. I. Metchkov, G. M. Mileva, and K. V. Stamenov, Phys. Lett. 89A, 441 (1982).

[6]Yu. I. Geller, V. F. Lukinykh, A. K. Popov, and V. V. Slabko, Opt. Spectrosc. (USSR) 51, 407 (1981); Phys. Lett. 82A, 4 (1981).

[7]U. Fano, Phys. Rev. 124, 1866 (1961).

[8]H. Klar and H. Kleinpoppen, J. Phys. B 15, 933 (1982); H. Kaminski, J. Kessler, and K. J. Kollath, Phys. Rev. Lett. 45, 1161 (1980).

[9]P. Lambropoulos, in *Advances in Atomic and Molecular Physics* (Academic, New York, 1976), Vol. 12. p. 87.

[10]We are not concerned with strong probe field effects discussed for autoionizing and autoionizinglike resonances in the following papers: L. Armstrong, Jr., C. E. Theodosiou, and M. J. Wall, Phys. Rev. A 18, 2538 (1978); P. Lambropoulos, Appl. Opt. 19, 3926 (1980); P. Lambropoulos and P. Zoller, Phys. Rev. A 24, 379 (1981); K. Rzazewski and J. H. Eberly, Phys. Rev. Lett. 47, 408 (1981); P. E. Coleman, P. L. Knight, and K. Burnett, Opt. Commun. 42, 171 (1982); P. E. Coleman and P. L. Knight, J. Phys. B 15, L235 (1982); G. S. Agarwal, S. L. Haan, K. Burnett, and J. Cooper, Phys. Rev. Lett. 48, 1164 (1982); G. Alber and P. Zoller, Phys. Rev. A (in press).

[11]U. Fano, Phys. Rev. 178, 131 (1969); 184, 250 (1969).

[12]P. Lambropoulos, Phys. Rev. Lett. 30, 413 (1973).

[13]S. N. Dixit, P. Lambropoulos, and P. Zoller, Phys. Rev. A 24, 318 (1981).