RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 40, NUMBER 11 DECEMBER 1, 1989

Adiabatic population transfer in a three-level system driven by delayed laser pulses

J. R. Kuklinski,* U. Gaubatz,† F. T. Hioe,‡ and K. Bergmann
Fachbereich Physik der Universität Kaiserslautern, Postfach 3049, D-6750 Kaiserslautern, West Germany
(Received 15 September 1989)

We give a simple analytic solution that describes a novel method for population transfer in a three-level system driven by delayed pulses and which accounts for recent experimental results. This solution describes a procedure that is counterintuitive, and yet it is shown to be, in fact, one of the simplest solutions for multilevel systems arising from the adiabatic theorem. Its possible application to many-level systems is suggested.

INTRODUCTION

The development of techniques for efficient transfer of population to thermally unpopulated atomic or molecular levels, such as high-lying Rydberg states or molecular vibrational levels, is of crucial importance to further advance our knowledge in spectroscopy and collision dynamics. It has been suggested[1] and independently experimentally demonstrated[2] that complete transfer of population can be achieved when atoms or molecules are exposed to two laser fields with an appropriate time dependence. In this Rapid Communication, we give a simple approximate analytic solution to this problem, which describes an efficient way for transfer of population into excited atomic or molecular levels that are not accessible by one-photon transitions. This solution is found to fit very well with the result of a recent experiment involving the excitation of sodium molecules by two spatially displaced laser beams.[2] In this experiment, the molecules interact first with the Stokes laser and then with the pump laser. Unlike other solutions[3] which usually depend sensitively on the input parameters such as the laser pulse shapes, intensity, and frequency modulation, the adiabatic following solution discussed by us is quite insensitive to changes in those parameters as long as certain easily controllable experimental conditions are satisfied. From the theoretical point of view, this solution is interesting not only for its remarkable simplicity, but also because (i) it prescribes a procedure that is counterintuitive, (ii) it is one of the simplest analytic solutions that describes the adiabatic rapid passage[4] for multilevel systems, a process of fundamental importance in quantum mechanics, and (iii) it generalizes a relation that expresses population trapping.[5] We mention that Peterson, Cantrell, and Burkey[6] have previously pointed out that multiphoton excitations of multilevel systems can be described more accurately using an adiabatic theorem than the sudden approximation for many experimental cases.

A SPECIFIC EIGENFUNCTION OF THE HAMILTONIAN

Consider a three-level system with eigenvectors $|1\rangle$, $|2\rangle$, and $|3\rangle$. We discuss the problem of transferring population, which is initially in level 1 to level 3 (see Fig. 1) by means of a pump laser which connects levels 1 and 2 and a Stokes laser which connects levels 2 and 3. The strength of the interaction varies with time and is given by the respective Rabi frequency $\Omega_n(t)=\mu E_n(t)/\hbar$, where μ is the relevant dipole matrix element and $E(t)$ the time-dependent amplitude of the corresponding laser field.

Under two-photon resonance condition and using the rotating-wave approximation, the Hamiltonian of the system can be written as[7]

$$\hat{H}=-\tfrac{1}{2}\hbar\begin{bmatrix}0 & \Omega_1(t) & 0\\ \Omega_1(t) & 2\Delta(t) & \Omega_2(t)\\ 0 & \Omega_2(t) & 0\end{bmatrix}, \tag{1}$$

where $\Delta(t)$ is the one-photon detuning of the laser frequencies from the respective transitions; see Fig. 1.

Using

$$\tan\theta(t)=\frac{\Omega_1(t)}{\Omega_2(t)}, \tag{2}$$

it is easy to verify that

$$|u_1(t)\rangle=\cos\theta(t)\,|1\rangle-\sin\theta(t)\,|3\rangle \tag{3}$$

is one of three eigenvectors of the Hamiltonian, Eq. (1), at any time t and independent of Δ. This eigenvector has a zero eigenvalue, independent of the Rabi frequencies. It is of particular interest because it is the only eigenvector which does not include a contribution of the intermediate level 2. We will show below under which conditions Eq. (3) is an adiabatic following solution, describing complete population transfer between levels 1 and 3.

We assume that only level 1 is initially populated.

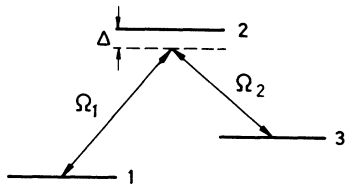

FIG. 1. Three-level system coupled by two lasers. The Rabi frequencies for the pump and Stokes laser are Ω_1 and Ω_2, respectively. The detuning from the transition frequency is Δ for both lasers, provided that the two-photon (Raman) resonance is maintained.

Complete population transfer occurs if

$$\left.\frac{\Omega_1(t)}{\Omega_2(t)}\right|_{t\to-\infty}\to 0 \text{ and } \left.\frac{\Omega_2(t)}{\Omega_1(t)}\right|_{t\to+\infty}\to 0, \quad (4)$$

where $t\to-\infty$ and $t\to+\infty$ corresponds to times before and after the interaction with the lasers, respectively, provided the evolution is adiabatic (see below). In fact, it follows from Eqs. (2)–(4) that we have

$$\begin{aligned} &|\langle 1|u_1(t)\rangle|^2=1 \text{ for } t\to-\infty, \\ &|\langle 3|u_1(t)\rangle|^2=1 \text{ for } t\to+\infty. \end{aligned} \quad (5)$$

Experimentally, Eq. (4) requires that the interaction of the Stokes laser with the atom or molecule begins and ends earlier than the interaction with the pump laser (see Fig. 2). This sequence seems to be counterintuitive. It can be achieved by appropriate spatial displacement of the axes of cw lasers when interaction with particles of a molecular beam is considered or by a suitable time delay of the pump laser relative to the Stokes laser when pulsed lasers are used.

CONDITIONS FOR ADIABATIC FOLLOWING

We now consider the conditions under which the system with the state vector

$$|\Psi(t)\rangle=|1\rangle \text{ for } t\to-\infty$$

[see Eq. (5)] evolves adiabatically, i.e., remains very nearly an eigenvector of the time-dependent Hamiltonian at all times. [$|\Psi(t)\rangle\approx|u_1(t)\rangle$].

Nonadiabatic coupling between the eigenstates is small when the rate of change of the mixing angle $\theta(t)$, Eq. (2), is small compared to the separation $\Delta\omega$ of the corresponding eigenvalues, see Messiah.[4] For $\Delta=0$, the latter are given by $\Delta\omega=\pm 0.5(\Omega_1^2+\Omega_2^2)^{1/2}=\Omega_{\text{eff}}$. Therefore $|d\theta/dt|\ll\Omega_{\text{eff}}$ is required for small nonadiabatic coupling.

It follows from Eq. (3) that

$$\dot\theta(t)=\frac{\dot\Omega_1(t)\Omega_2(t)-\Omega_1(t)\dot\Omega_2(t)}{\Omega_1^2(t)+\Omega_2^2(t)}. \quad (6)$$

We introduce

$$\Omega_{1,2}(t)=\Omega_{1,2}^0 g(\tau), \quad (7a)$$

and use, for convenience,

$$g_{1,2}(\tau)=\exp[-(\tau+\delta_{1,2})^2], \quad (7b)$$

where $\tau=t/T$ and $\delta_{1,2}=\mp\Delta t/T$ measure the time and time delay (or displacement), respectively, in units of the pulse length (or interaction time) T. The + and − signs apply to the Stokes laser [$\Omega_2(t)$] and the pump laser [$\Omega_1(t)$], respectively. With Eqs. (6) and (7), together with the assumption that the delay is of the order of the pulse width, i.e., $|\delta|\approx 1$, the adiabatic theorem leads to the condition [for further details see Ref. (8)]

$$\Omega_{\text{eff}}T\gg 1. \quad (8)$$

It is a nontrivial consequence of Eq. (8) that $T\to\infty$ is not required to reach the asymptotic limit of adiabatic following. Rather, for given T, the limit can be approached by increasing Ω_{eff}. This is exactly the point of interest for experiments with intense laser, since it shows that the adiabatic limit can be achieved for strong enough pulses even if the pulse duration is short.

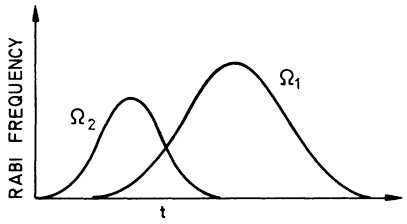

FIG. 2. Time dependence of the Rabi frequencies required for efficient population transfer under adiabatic following conditions.

Alternatively, we may consider the Schrödinger equation scaled to the dimensionless parameter τ, and using for convenience $\Omega_1^0=\Omega_2^0=\Omega^0$,

$$\frac{1}{\Omega_0 T}\frac{d}{d\tau}|\Psi(\tau)\rangle=-\frac{i}{\hbar}\hat H(\tau)|\Psi(\tau)\rangle, \quad (9)$$

with

$$\hat H(\tau)=\hbar\begin{bmatrix} 0 & g_1(\tau) & 0 \\ g_1(\tau) & 2\Delta(\tau)/\Omega^0 & g_2(\tau) \\ 0 & g_2(\tau) & 0 \end{bmatrix}. \quad (10)$$

Obviously, when Eq. (8) is satisfied, the regime of adiabatic following is reached. Then Eq. (9) reduces to

$$\hat H(\tau)|\Psi(\tau)\rangle=0 \quad (11)$$

at all times τ. Thus, steady-state condition is maintained at any time throughout the process.[1,9] If the system is initially (at $t\to-\infty$) prepared in the eigenstate $|u_1\rangle$ it remains in that eigenstate at all times and Eq. (3) is in fact the adiabatic following solution for complete population transfer from level 1 to level 3.

A more rigorous treatment of the adiabatic following conditions, for on-resonance excitation $\Delta=0$ (see, e.g., Ref. 8), leads, instead of Eq. (8), to

$$\frac{1}{\Omega^0 T}\frac{|g_1(\tau)\dot g_2(\tau)-\dot g_1(\tau)g_2(\tau)|}{[g_1^2(\tau)+g_2^2(\tau)]^{3/2}}\ll 1, \quad (12)$$

where the dot identifies the derivative with respect to τ. Equations (8) or (12) implies that details of the pulse shape are not important. It is also obvious from Eq. (8) [or Eqs. (7) and (12)] that the overlap of the interaction with the two lasers should not be too small. For $\delta\gg 1$ the denominator of Eq. (12) (or $\Omega_{\text{eff}}^2=\Omega_1^2+\Omega_2^2$) may become very small and Eq. (8) [or Eq. (12)] is no longer easily satisfied.

The discussion presented in the Schrödinger picture can be extended to the Heisenberg picture. In this case, we deal with the Liouville equation for the reduced density matrix

$$\frac{1}{T}\frac{d\rho}{d\tau}=\frac{i}{\hbar}[\rho,H]+L\rho. \quad (13)$$

Using this approach, the analysis can be further extended to include relaxation processes during the evolution of the population transfer, given by the matrix elements L_{ik}. Radiative and collisional damping, e.g., both contribute to $L_{22}=\gamma_{22}$, while γ_{13} is the relaxation of the Raman coherence.

The adiabatic following solution discussed above is, of course, given by $\rho_{ad}(\tau)=|u_1(\tau)\rangle\langle u_1(\tau)|$.

We can verify by inspection that the same solution as in the lossless case $L=0$ arises, provided Eqs. (4) and (8) are satisfied and $\gamma_{ik}T\ll 1$ is valid, except for γ_{22}. Restrictions for the matrix element L_{22} need not be invoked, because it does not affect the eigenstate $|u_1\rangle$ or its eigenvalue.

We emphasize the most important conclusion, namely, the independence of the transfer efficiency from radiative or collisional damping of the intermediate level 2. In fact, it has been noted previously by Hioe and Carroll[10] that in general the process of an adiabatic rapid passage or adiabatic following tends to minimize the population of the intermediate level 2 in a three-level system. A related analysis in which concurrent instead of delayed pulses were used but in which the one-photon detuning $\Delta(t)$ was varied in time was given by Oreg, Hioe, and Eberly.[1] These authors also suggested a counterintuitive way of varying the one-photon detuning in order to achieve efficient population transfer. The result derived in this paper, however, gives the simplest analytical form which also reveals the most essential features clearly and explicitly.

Finally, we consider the consequences of having the laser frequencies not tuned to exact two-photon resonances, i.e., when $\Delta_1(\tau)\neq\Delta_2(\tau)$. In that case, the matrix element in the lower right corner of the Hamiltonian, Eq. (10), is not zero anymore. It is rather given by $[\Delta_1(\tau)-\Delta_2(t)]/\Omega^0$. Again, we realize that the adiabatic following solution is approximately valid as long as the detuning from the two-photon resonance is small compared to the Rabi frequency Ω^0 (or Ω_{eff}). We also note that phase fluctuations of the lasers during the interaction time with the molecules are assumed to be negligibly small. This is appropriate for the interaction of molecules with single-mode cw lasers.

TRAPPED STATE

The fact that $|u_1(t)\rangle$ is an eigenvector corresponding to the zero eigenvalue makes this solution a special generalization of the relation for population trapping, as we shall briefly explain below.

To the best of our knowledge, population trapping has been observed and discussed[5,11] for constant laser fields, $\Omega_{1,2}=\text{const}$, or concurrent pulses $\Omega_1(t)=c\,\Omega_2(t)$, only. For these cases it follows from Eqs. (2), (3), and (7) that $|u_1\rangle$ is a constant eigenvector because the mixing angle θ is time independent. A statement expressing population trapping is[5]

$$\cos\theta\langle\Psi(t)|1\rangle-\sin\theta\langle\Psi(t)|3\rangle=\text{const}. \tag{14}$$

The above relation also holds for the adiabatic following process where θ is time dependent. To prove this statement, we realize that we have from $i\hbar(\partial/\partial t)|\Psi(t)\rangle=\hat{H}(t)|\Psi(t)\rangle$,

$$i\hbar\langle u_1|\partial/\partial t|\Psi(t)\rangle=\langle u_1|\hat{H}(t)|\Psi(t)\rangle=0, \tag{15}$$

because $\langle u_1|\hat{H}(t)=0$. If $\langle u_1|$ is time independent, as in the case of concurrent pulses, the left-hand side of Eq. (15) can be written as $i\hbar\partial/\partial t\langle u_1|\Psi(t)\rangle$, and hence $\langle u_1|\Psi(t)\rangle=\text{const}$, the explicit statement of population trapping, follows. When $\langle u_1|$ is slowly time varying in the sense prescribed above Eq. (6) for the adiabatic following process, the left-hand side of Eq. (15) can be approximately written as $i\hbar(\partial/\partial t)\langle u_1|\Psi(t)\rangle$ also. Thus, our approximate analytical solution for the adiabatic process also gives a generalization of the previously known relation for population trapping.

OUTLOOK AND SUMMARY

An extension of the above analysis and solution to a system of more than three levels is straightforward. In the case in which several levels and several stepwise transitions of, say, decreasing level spacings are involved, such as in the case of, e.g., levels in hydrogenic atoms or vibrational levels in an anharmonic potential, the process of excitation can be suitably replaced by one that changes continuously the detuning. Efficient population transfer can be accomplished by either tuning the frequency of the incident laser pulse or by appropriate Stark shifting the levels by a varying external electric field in such a way that the interaction is first between the uppermost pair of levels and last with the lowermost one, still partially overlapping in time with the former. This is again a counterintuitive interaction sequence. Such processes can be used, for example, for the efficient production of hydrogen atoms in the high angular momentum states, the so-called circular states.[12] It may also be relevant for multiphoton excitation and dissociation of molecules as was recently demonstrated by Liedenbaum, Stolte, and Reuss.[13] A similar multiphoton-absorption technique may also prove to be quite efficient for the ionization of certain atoms or molecules.

In summary, we have found a remarkably simple approximate solution of the Schrödinger equation describing the evolution of the state vector for a three-level system interacting with two radiation fields while the two-photon resonance is maintained. It accounts for the experimentally observed efficient population transfer.[2] A detailed account of this experiment,[8] as well as a detailed numerical study to assess the prospect of applying this technique with continuous or pulsed lasers to a large class of molecules,[14] will be published elsewhere.

ACKNOWLEDGMENTS

This work was the result of a collaborative effort during a visit of two of us (J.R.K. and F.T.H.) to Kaiserslautern. Support from the Deutsche Forschungsgemeinschaft under Grant No. SFB 91 "Energietransfer in atomic and molecular collisions" is gratefully acknowledged. One of

us (F.T.H.) would like to thank the U.S. Department of Energy, Division of Chemical Sciences for support, and also Dr. C. E. Carroll and Professor J. H. Eberly for many useful discussions. J.R.K. was partially supported by the Polish Government Grant No. CPBP01.07.

---

*Permanent address: Institute of Theoretical Physics of the Academy of Sciences, Warsaw, Poland. Present address: University of Rochester, Department of Chemistry, River Station, Rochester, NY 14627.

†Present address: Department of Physics, State University of New York, Stony Brook, NY 11794-3800.

‡On leave from the Department of Physics, St. John Fisher College, Rochester, NY 14618; also with: Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627.

[1]F. T. Hioe, Phys. Lett. **99A**, 150 (1983); J. Oreg, F. T. Hioe, and J. H. Eberly, Phys. Rev. A **29**, 690 (1984).

[2]U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988).

[3]See, e.g., C. E. Carroll and F. T. Hioe, J. Math. Phys. **29**, 487 (1988).

[4]A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1962), Vol. II, p. 744.

[5]G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cimento B **36**, 5 (1976); E. Arimondo and G. Orriols, Lett. Nuovo Cimento **17**, 333 (1976); H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978); R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A **14**, 1498 (1976); S. Swain, J. Phys. B **15**, 3405 (1982); P. M. Radmore and P. L. Knight, *ibid.* **15**, 561 (1982); F. T. Hioe and C. E. Carroll, Phys. Rev. A **37**, 3000 (1988).

[6]G. L. Peterson, C. D. Cantrell, and R. S. Burkey, Opt. Commun. **43**, 123 (1982).

[7]See, e.g., F. T. Hioe, Phys. Rev. A **29**, 3434 (1984).

[8]U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann (unpublished).

[9]For another approximate solution for adiabatic rapid passage, see also D. Grischkowsky, M. M. T. Loy, and P. F. Liao, Phys. Rev. A **12**, 2514 (1975).

[10]F. T. Hioe and C. E. Carroll, in Proceedings of the Sixth Rochester Conference on Coherence, Rochester, 24–31 June 1989 (unpublished).

[11]M. Kaivola, N. Bjerre, O. Poulson, and J. Javanainen, Opt. Commun. **49**, 418 (1984); R. J. McLean, R. J. Ballagh, and D. M. Warrington, J. Phys. B **18**, 2371 (1985).

[12]R. G. Hulet and D. Kleppner, Phys. Rev. Lett. **51**, 1430 (1983); J. A. Yeazell and C. R. Stroud, Jr., *ibid.* **60**, 1494 (1988).

[13]C. Liedenbaum, S. Stolte, and J. Reuss, Phys. Rep. **178**, 1 (1989).

[14]U. Gaubatz, S. Schiemann, A. Kuhn, and K. Bergmann (unpublished).