# Efficient polarization of high-angular-momentum systems

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## ABSTRACT

We present a novel technique of efficient optical pumping of open, high-angular-momentum systems. The method combines two well-established approaches of population manipulation (conventional optical pumping and coherent population transfer), offering the ability to achieve higher population of a sublevel with the highest or lowest quantum number $m$ (the "end state") than obtainable with either of the techniques. To accomplish this task, we propose to use coherent-population-transfer technique (e.g., adiabatic fast passage) to arrange the system in such a way that spontaneously emitted photon (conventional optical pumping) carries away more entropy than in conventional schemes. This enables reduction of a number of spontaneous decays $N_{\rm sd}$ required to pump the system with the total angular momentum $J$ from $N_{\rm sd} = J$ decays in the conventional scheme to $N_{\rm sd} \lesssim \log_2(2J)$ decays in the proposed scheme. Since each spontaneous-emission event is potentially burdened with a loss of population (population is transferred to a dark state), this enables increasing population accumulated in the "end state", which is important for many applications.

## 1. INTRODUCTION

A stretched state, i.e., a state in which only the Zeeman sublevel with the highest or lowest magnetic quantum number $m$ (the "end sublevel") is populated, plays an important role in many quantum optics, atomic physics (see, for example, Ref.[1] and references therein), and metrological experiments.[2–4] Due to their purity, the states often serve as a starting point for quantum-information protocols or are used in manipulations of quantum systems.[5,6] Particularly, controlling interaction of magnetic moments of particles pumped into the stretched state with spatially varying magnetic fields enables cooling of gases to ultra-low temperatures (magneto-optical cooling).[7] Moreover, due to the dependence of some chemical reactions on the relative polarization of two interacting compounds, pumping the system into the stretched state offers the ability to control dynamics of the reactions (see, for example, Ref.[8] and references therein).

In addition to the purity of the stretched state, the state possesses another interesting property – it is immune to some relaxation mechanisms (e.g., spin-exchange-collision relaxation).[9,10] Since in many systems (e.g., atomic vapors at room or alleviated temperatures) such relaxation limits the lifetime of quantum states, restraining the relaxation allows the prolongation of systems' lifetimes. This results in the appearance of narrow spectral features in signals detected with light interacting with the systems.[11–13] Specifically, elimination of spin-exchange relaxation via pumping into the stretched state has recently enabled ultra-sensitive (femtotesla level) detection of external magnetic fields (see, for example, Ref.[4] and references therein).

The stretched state can also be used in efficient generation of spin squeezing. This is performed via process of orientation-to-alignment conversion,[14] in which an electric field of light reduces of spin-projection uncertainty in a certain direction below the standard limit, i.e., limit obtained with coherent states,[15] at the expanse of

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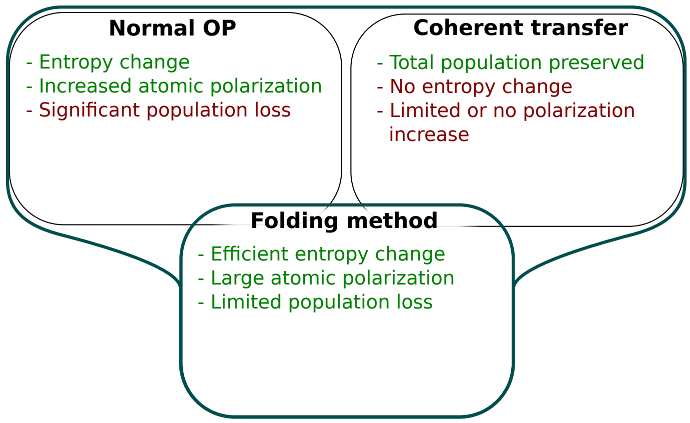


Figure 1. Advantages and disadvantages of conventional optical pumping and coherent-population-transfer schemes in generation of the stretched state in high total-angular-momentum system. The folding scheme combines attributes of these two approaches, offering the ability to efficently pump atoms into the "end sublevel" without significant deterioration of total population of the pumped level.

uncertainty in another direction. Such states are particularly interesting as spin-squeezing may offer enhanced sensitivity in precise measurements, such as magnetometry (see Ref.[16] and references therein).

While optical pumping (OP) of simple, low total-angular-momentum systems, such as alkali atoms, is relatively straightforward, pumping systems with high angular momentum (e.g., molecules with a large rotational quantum number $J$) is a significantly more challenging. In this paper, we propose a novel technique of stretched-state generation, enabling efficient pumping of high-angular-momentum systems. The technique makes use of two well-known approaches (conventional optical pumping and coherent population transfer), providing the new approach with capabilities to transfer population with efficiency significantly exceeding that of conventional techniques. This enhanced efficiency may find particular applications in generation of spin squeezing via orientation-to-alignment conversion, as such a technique works best with the system of high total angular momentum.[14]

## 2. CONVENTIONAL OPTICAL PUMPING VS. COHERENT-POPULATION-TRANSFER TECHNIQUES

Conventional optical pumping (COP) of atoms/molecules requires many cycles of photon absorption (from circularly polarized light) and subsequent spontaneous emission to transfer population into the "end sublevel" (generation of the stretched state). While during this process significant atomic polarization may be achieved, the potential problem with this scenario may be deterioration of population of the light-coupled ground state; unless closed transition is employed, optically excited particles can relax into other long-lived levels, becoming "invisible" to light and hence lost. In the worst-case scenario, particles can be fully polarized in a given level (whole population of the level is contained in the "end sublevel"), while the population of the "end sublevel" is practically unaffected with light (the other sublevels are depleted). This may exclude the scheme from many applications. Figure 1 summarizes the most important features of COP in the context of pumping of high angular-momentum systems.

An interesting alternatives to COP are coherent-population-transfer (CPT) techniques. The techniques enable transferring population between two quantum states without the necessity of application of spontaneous emission. A seminal example of such techniques is adiabatic-fast-passage (AFP) technique, which swap populations of two states by application of frequency-chirped optical pulses. Another approach is stimulated Raman adiabatic passage (STIRAP), which uses a two-pulse sequence to transfer population from a populated ground state to an unpopulated state with an aid of an intermediate state. The most important feature of this scheme is elimination of appreciable excitation of the intermediate state,[17] which enables application of the technique in systems characterized with a short-lived intermediate (excited) state. The problem, however, with all CPT techniques is that, by themselves, the techniques cannot be used to combined population of various sublevels into the "end sublevel". Advantages and disadvantages of this technique are shown in Fig. 1.

To understand the difference between COP and CPT methods, one can consider the change of entropy associated with pumping of a quantum system into the stretched state. Based on theoretical considerations, it can be shown that the von Neumann entropy per atom $S_{\text{at}}$ of the system in the stretched state is zero ($S_{\text{at}} = 0$), while the entropy of the system with equally populated sublevels, corresponding to thermal equilibrium, is maximal.[18] Thus, OP into the stretched state is associated with the change of system's entropy. In COP, the entropy difference between the initial and final states is carried away by spontaneously emitted photons. This can be shown by the non-Hermiticity of spontaneous-emission operator, denoting irreversibility of the relaxation process. On the other hand, the operator describing CPT processes are unitary (the processes are reversible). Since unitary transformations cannot change the eigenvalues of a matrix they act on, application of the CPT operators to the density matrix describing the initial (thermal-equilibrium) state of the system cannot change the population of the latter (entropy is preserved).

While CPT techniques alone cannot be used to created the stretched state (other than by simply depleting all sublevels except for the "end sublevel", which is not better than the worst-case efficiency of COP), they can be used to enhance the efficiency of optical pumping (see, advantages of the folding technique shown in Fig. 1). This increase stems from the fact that COP uses far more spontaneous-decay events per atom than necessary to convert the initial entropy of the system into its final entropy. In such a case, the combination of coherent and dissipative processes enables optimization of entropy carried away by spontaneously emitted photons and hence reduces the number of spontaneous decays.

## 3. FOLDING METHOD OF OPTICAL POLARIZATION

For the sake of discussion of the folding scheme, we consider a generic system $J \to J'$ with an initially unpolarized ground state (Fig. 2). During the $1^{\text{st}}$ pumping cycle, the system interacts with a sequence of $J$ alternating AFP pulses of orthogonal circular polarizations to transfer population from the $m < 0$ ground-state sublevels to the $m' > 0$ excited-state sublevels (Fig. 2). To avoid uncontrollable losses during the stage, the sequence must be completed within the lifetime of the excited state. After sequence completion, about half of population resides in the excited state. These atoms subsequently decay into already populated ground-state sublevels, approximately doubling populations of the sublevels*. During the $2^{\text{nd}}$ cycle, the sequence is repeated with $\approx J/2$ pulses and it is followed by waiting time allowing for spontaneous emission to occur. This pumping cycle increases populations of the $m \gtrsim J/2$ sublevels on the expanse of the $m < J/2$ sublevels. In general, in each cycle the number of AFP pulses is reduced in half with respect to the former cycle (during the $i^{\text{th}}$ pumping stage approximately $J/2^{(i-1)}$ pulses is applied). The transfer-decay procedure is repeated until whole population of the $J$ level is pumped into the "end sublevel" (and potentially into the closest sublevel†). To pump atom into the stretched state, approximately $\log_2 2J$ iterations is required‡.

In case of the closed system, where upper state only relaxes into the state it was excited from, the pumping scheme will eventually transfer whole population into the "end sublevel". If, however, the atoms can relax into other states (the branching ratio $R$ is smaller than unity, $R < 1$), some population will be lost during the process. The final population of the "end sublevel" can be estimated as follows. The initial population of the sublevel is $1/(2J+1)$ and in each iteration it is multiply approximately by $1 + R$. Assuming $\log_2(2J)$ pumping cycles to polarize the system, we obtain the formula on the final population of the "end sublevel" $\rho^{\text{folding}}_{Jm=J,Jm=J}$

$$\rho^{\text{folding}}_{Jm=J,Jm=J} \approx \frac{(1+R)^{\log_2(2J)}}{2J+1}. \tag{1}$$

*Increase of populations in the ground-state sublevels strongly depends on the branching ratio from the excited to the ground state.

†Depending on the actual energy structure of the system.

‡To further increase population of the "end sublevel", additional COP light pulses can be applied.

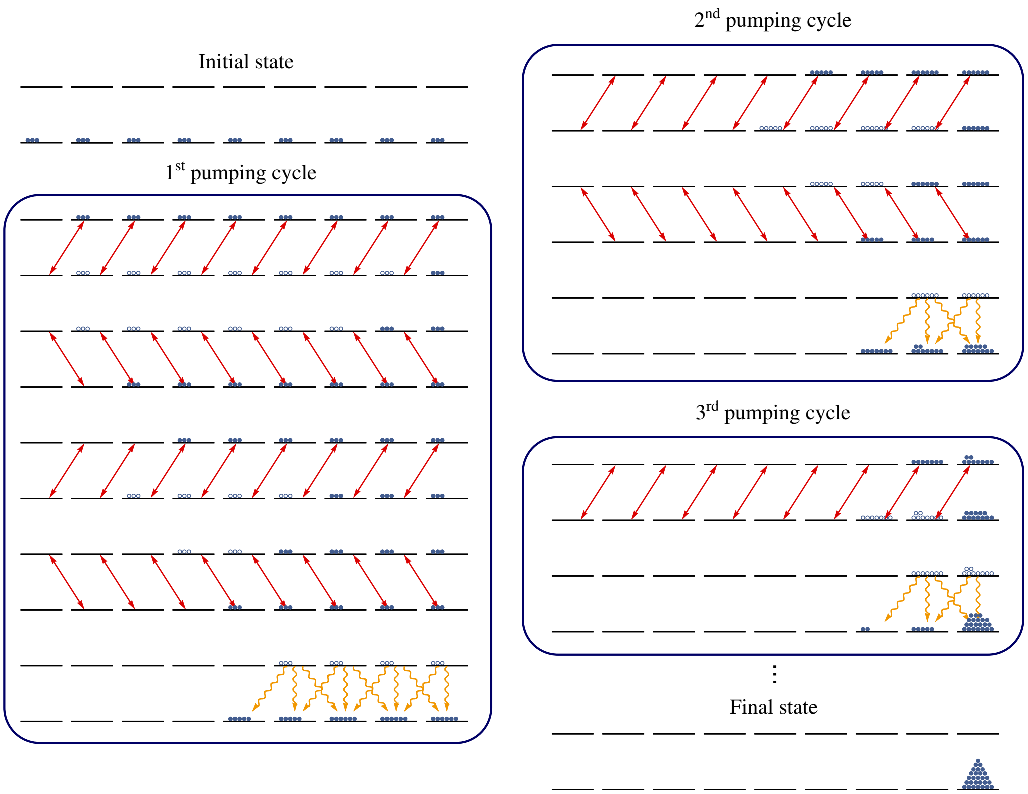


Figure 2. Schematic of the folded method used for efficent optical pumping to the $m = J$ sublevels in an exemplary $J = 4 \rightarrow J' = 4$ system. Population before each pulse is shown as open circles, and after each pulse as closed circles. The first diagram shows the initial state with unpolarized ground state and unpopulated upper state. Three folding procedures are applied, each followed by spontaneous decay of the upper state. In the $1^{\text{st}}$ cycle, four ($= J$) AFP pulses are applied, with polarizations $\sigma^+$, $\sigma^-$, $\sigma^+$, $\sigma^-$. In the $2^{\text{nd}}$ cycle, two ($= J/2$) pulses are applied, with polarizations $\sigma^+$, $\sigma^-$, and during the $3^{\text{rd}}$, one ($= J/2^2$) $\sigma^+$ pulse is applied. This sequence can be followed by several cycles of conventional optical pumping to further increase the "end-sublevel" population.

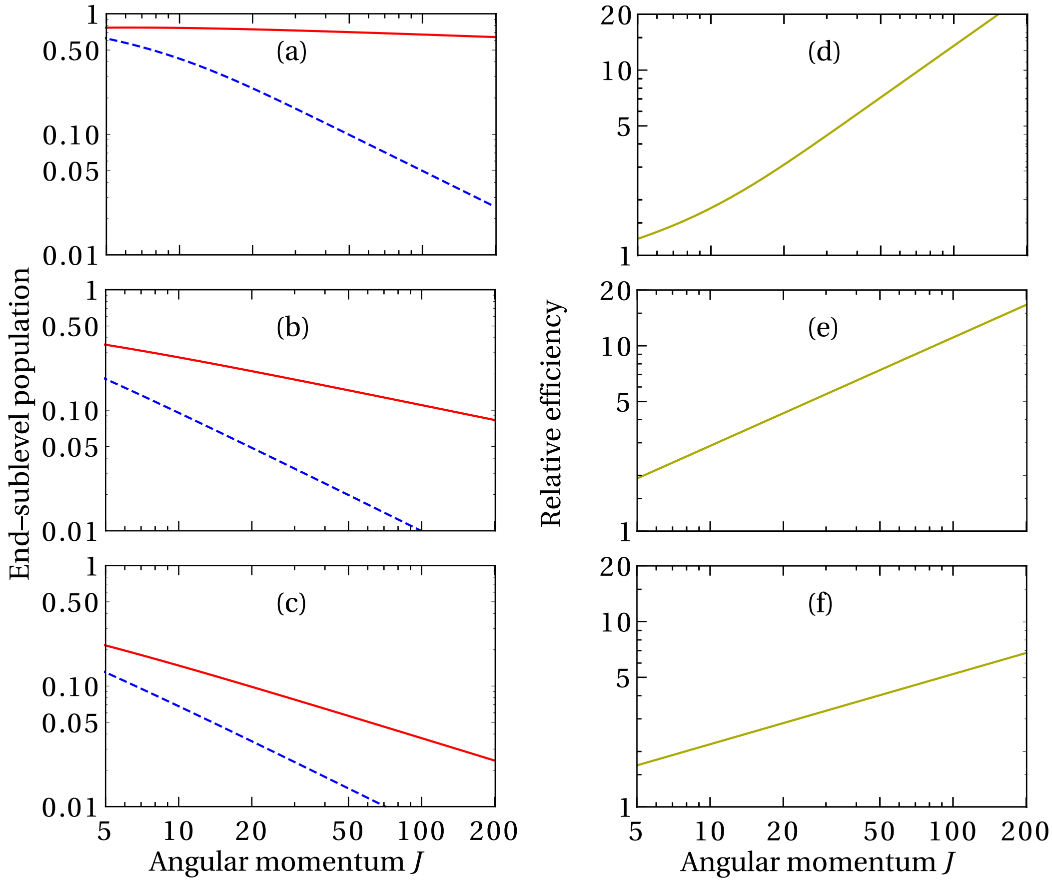


Figure 3. Population of the "end sublevel" obtainable with the folding method (solid red line) and conventional optical pumping (dashed blue line) versus the total angular momentum $J$ [left column; (a)-(c)] and ratio of the efficiency of optical pumping with the two techniques [right column; (d)-(f)]. The calculations were performed in an open system $J \to J' = J$ with three excited-state branching ratios $R$: (a) and (d) $R = 0.9$, (b) and (e) $R = 0.5$, and (c) and (f) $R = 0.3$. The curves show estimates given by Eqs. (1) and (2).

In conventional optical pumping, the transfer of population from the $m$ sublevel to the "end sublevel" requires about $J - m$ spontaneous-decay events[§]. Summing over all sublevels, one can estimate the final population of the "end sublevel" $\rho^{\rm COP}_{Jm=J,Jm=J}$

$$\rho^{\rm COP}_{Jm=J,Jm=J} \approx \frac{1}{2J+1}\sum_{m=-J}^{J} R^{J-m} = \frac{1-R^{2J+1}}{(2J+1)(1-R)}. \tag{2}$$

This equation, in conjunction with Eq. (1) can be used to compare efficiency of optical pumping with conventional and our schemes.

Figure 3 compares efficiency of optical pumping (the population of the "end sublevel") versus the total angular momentum $J$ for the folding scheme and conventional optical pumping. In both schemes, relaxation of the excited state to a different ground state than the one particle was excited from ($R < 1$) results in population depletion, i.e., final population of the "end-sublevel" is smaller than unity. Since pumping of higher total-angular-momentum

[§]In our considerations, we neglect the difference in the Clebsch-Gordan coefficients, which shows different trends depending on the structure. This difference may affect efficiency of optical pumping, particularly in COP, but it does no fundamentally modify the process.

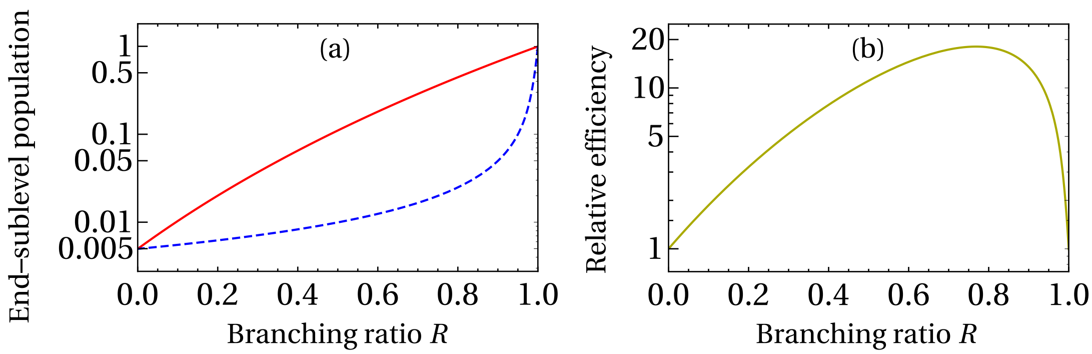


Figure 4. Final population of the "end sublevel" achieved with folding scheme (solid red line) and conventional optical pumping (dashed blue line) (left) and ratio of the population obtainable with these techqniques (right) as a function of the branching ratio $R$. The calculations were performed based on Eqs. (1) and (2) in the $J = 100 \rightarrow J' = 100$ system.

system requires more pumping cycles and hence more spontaneous decays, final population of the sublevel decreases with $J$. The population deterioration is less pronounced in the folding scheme than in the COP scheme. Particularly, in a system with branching ratio $R = 0.9$, nearly 70% of the total population of the $J = 100$ level is transferred to the "end sublevel". This is approximately 13 times higher population than population of the "final sublevel" obtainable with the COP scheme ("end-state" population is about 5% of initial population of a whole level). As shown with other plots in Fig. 3, this difference depends on the branching ratio $R$, but in all cases the efficiency of the folding scheme is from several to several tens of times larger in the efficiency of COP.

Figure 4 presents the final population of the "end sublevel" on the branching ratio $R$. Both techniques lead to the same population of the "end sublevel" in two cases, i.e., for $R = 1$ ($\rho_{Jm=J,Jm=J} = 1$) and $R = 0$ ($\rho_{Jm=J,Jm=J} = 0$). In all other cases, the efficiency favors the folding scheme. The ratio reaches its maximum for the systems with the branching ratio $R_{\max} \approx 0.8$. It can be shown that the difference in efficiency increases with $J$ (while the "end-sublevel" population decreases in both cases). The increase is also accompanied by a shift of the value of the branching ratio that maximizes the efficiency ratio.

One of the problems in implementation of the folding scheme may be short lifetime of the excited state particles are pumped to. Short lifetime is problematic as whole pulse sequence needs to be completed within the lifetime of the state. In such a case, one may consider application of other CPT techniques. Particularly, additional long-lived state may enable application of STIRAP so that no population is generated in short-lived excited state. This alleviates a limitation on the timing and power of pulses in folding scheme, which may simplify practical implementations of the scheme. Moreover, the STIRAP scheme may also offer the ability to enhance the efficiency of the process. Particularly, with another additional long-lived (shelving) states, i.e., in a system with three long-lived levels and one short-lived level, one can transfer population into the "end state" with only one spontaneously emitted photon. This is the fewest to generated the stretched state that is enabled by the second law of thermodynamic. The detailed description of the schemes will be provided in the forthcoming publication.[19]

## 4. CONCLUSION

In this paper, we have described the new scheme of optical pumping, enabling efficient generation of the stretched state in high-angular-momentum systems, such as diatomic molecules in states with high rotational excitation. The improvement on the efficiency of optical pumping is accomplished by combination of conventional optical pumping and coherent-population-transfer scheme. In this scenario, prior to each spontaneous emission, population distribution is arranged with an aid of AFP pulses, folding population in half in each cycle. This optimizes entropy carried away by spontaneously emitted photos and hence reduces the number of spontaneous-emission events. The spontaneous decays transfer system to the ground state they were originally present, but also may also pump it into the state "invisible" to light, in which the particles are lost. With the folding approach we minimize the number of spontaneous-emission events as much as allowed by the second law of thermodynamics.

The use of such and similar processes enables the proposed method to be generalized beyond just populating the "end sublevel". In fact, once the population has been combined in one state, it can be moved, with and appropriate sequence of stimulated processes, to any other stage, or a coherent superposition thereof.

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