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Optimal conversion of an atomic to a molecular Bose-Einstein condensate

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The work in this article extends the optimal control framework of variational calculus to optimize the conversion of a Bose-Einstein condensate of atoms to one of molecules. It represents the derivation of the closed form optimal control equations for a system governed by a nonlinear Schrödinger equation and its successful application. It was necessary to derive a density matrix formulation of the coupled Gross-Pitaevskii equations to optimize STIRAP-like Raman light fields, to overcome dissipation.

Many-particle quantum dynamics reduced to effective one-particle quantum motion often give rise to nonlinear Hamiltonians and Schrödinger equations. An example is the Gross-Pitaevskii (GP) equation, which provides a highly successful framework for describing Bose-Einstein condensates (BECs) based on mean field theory. Recently these equations have been extended to treat the conversion of a BEC of atoms to a molecular BEC (MBEC) using external magnetic and light fields [1]. This method builds on the experimental successes achieved in creating an atomic condensate, by transforming that coherently into a condensate of molecules in a stable rovibrational ground state. The process makes use of a Raman pulse pair inducing the free-bound transition via an excited electronic state, combined with a sweep of the external magnetic field over a Feshbach resonance to enhance the total transition probability. The molecules are then translationally and rovibrationally cold. The conversion rate depends critically on the Raman fields used, which in previous work were all hand optimized guided by physical intuition [1–6]. In this article the required fields are calculated by optimal control theory [7,8], which is very successful in finding solutions close to the global optimum.

The BEC to MBEC conversion is, moreover, a challenge, because it is part of the important class of problems in which the coherent transfer is affected by dissipation on the same time scale. STIRAP [9] (stimulated Raman scattering involving adiabatic passage) solutions are in this case ideal candidates, overcoming dissipation by avoiding population in decaying levels. These counterintuitive pulse sequences are, however, not solutions of the closed form, rapidly convergent optimal control theory formulation based on mere population evolution [7,8]. Therefore there have been several attempts to devise other optimal control schemes, like local control [10] or gradient-type [11,12] optimizations, to naturally include these counterintuitive solutions. These methods, however, lack the global search capability of the closed form expressions and are therefore inferior. In this article the quantum control framework for the Liouville equation [13,14] is extended to nonlinear density matrix equations. This allows optimization of a STIRAP sequence of pulses to enhance the conversion rate from BEC to MBEC by a factor of 2 over previously published results [1].

The coupled Gross-Pitaevskii equations of Ref. [1] describe the evolution of a mixed atomic/molecular BEC under the influence of a Raman pulse pair and an external magnetic field. The atomic condensate (of Na atoms as an example) is described by the field \( \phi_1(\vec{x},t) \); the atom pairs in their intermediate Feshbach \( S+S \) quasibound state form a molecular Bose-Einstein condensate with order parameter \( \phi_3(\vec{x},t) \). Two further condensate components are considered: the molecules in the intermediate electronically excited \( S+P \) bound state (0\(_e\) symmetry) of the coherent Raman transition and the molecules in the final state, described by \( \phi_2(\vec{x},t) \) and \( \phi_3(\vec{x},t) \), respectively. The final internal state associated with \( \phi_3(\vec{x},t) \) is chosen to be the rovibrational ground state of the molecule in the Na+Na triplet potential. The amplitudes of the coherent fields are assumed uniform, \( \phi_{ij} = \sqrt{n_j} \exp(i \theta_j) \), and the interaction with the light fields with center frequencies \( \omega_1 \) and \( \omega_2 \) is taken in the rotating-wave approximation.

In this article we derive the density matrix analog of the GP equations of Ref. [1] to include also the evolution of the coherences in the system. The density matrix elements are \( \rho_{ij} = \langle \phi_i | \rho | \phi_j \rangle \). (This matrix elements describe the association of two atoms \( \phi_i \) into one molecule \( \phi_m \), and \( \rho_{ij} = \langle \phi_i | \rho | \phi_j \rangle \), with \( i,j = 1,...,3 \) indexing the molecular levels. Then the coupled Gross-Pitaevskii equations take the form where the populations are

\[
\begin{align*}
\dot{\rho}_a &= 2 \alpha \rho_{a1} - c.c., \\
\dot{\rho}_{11} &= E_1 \rho_{11} - \alpha \rho_{a1} + \Omega L_1 \rho_{12} - c.c., \\
\dot{\rho}_{22} &= E_2 \rho_{22} - \Omega L_1 \rho_{12} + \Omega L_2 \rho_{23} - c.c., \\
\dot{\rho}_{33} &= E_3 \rho_{33} - \Omega L_2 \rho_{23} - c.c., \\
\end{align*}
\]

and the coherences are

\[
\begin{align*}
\dot{\rho}_{12} &= (E_1 - 2 U_0 \rho_a) \rho_{12} - 4 \alpha \rho_a \left( \rho_{11} - \frac{1}{4} \rho_a \right) + \Omega L_1 \rho_{a2},
\end{align*}
\]
\[ i \dot{\rho}_{a2} = (E_2 - 2U_0 \rho_a) \rho_{a2} - 4 \alpha \rho_a \rho_{12} + \Omega_{L1} \rho_{a1} + \Omega_{L2} \rho_{a3}. \]
\[ i \dot{\rho}_{a3} = (E_3 - 2U_0 \rho_a) \rho_{a3} - 4 \alpha \rho_a \rho_{13} + \Omega_{L2} \rho_{a2}, \]
\[ i \dot{\rho}_{12} = (E_2 - E_1^*) \rho_{12} - \alpha \rho_{a2} + \Omega_{L1} (\rho_{11} - \rho_{22}) + \Omega_{L2} \rho_{13}, \]
\[ i \dot{\rho}_{13} = (E_3 - E_1^*) \rho_{13} - \alpha \rho_{a3} - \Omega_{L1} \rho_{23} + \Omega_{L2} \rho_{12}, \]
\[ i \dot{\rho}_{23} = (E_3 - E_2^*) \rho_{23} - \Omega_{L1} \rho_{13} + \Omega_{L2} (\rho_{22} - \rho_{33}). \]

The system described by these equations is essentially a Λ-type molecular system, coupled to a source of atoms via tunneling. \( U_0 \) is the off-resonant strength of the atomic condensate self-energy, \( \alpha \) the rate constant of atom to quasi-bound molecule conversion. The energy of the quasibound state \( E_i = e_i - (i/2) \gamma_0 \) is assumed complex, since it can dissociate into atoms leaving the trap, depending on the energy of \( e_i \) which is controlled by the external magnetic field. For \( e_i < 0 \) it is a bound molecular level and it cannot dissociate; thus \( \gamma_0 = 0 \). The magnetic field sweep is such that the real part of \( E_i \) undergoes a Zeeman shift varying linearly in time and crosses the resonance value \( 0 \) at time \( 0 \), at which instant the \( \rho_a \) to \( \rho_{11} \) conversion is most efficient. \( E_2 = e_2 - (i/2) \gamma_{sp} - \omega_{L1} \) is the complex (dressed) excited state energy with \( \gamma_{sp} \) the spontaneous decay width and \( E_3 = -\omega_{L1} + \omega_{L2} \) the energy of the final bound molecular state. For a more detailed description see Ref. [1]. Included in this model is the boson stimulation [6] and also the change of self-energy of the condensate due to reduction of the atomic BEC component during the conversion process. Not included is the effect of atom-molecule and molecule-molecule self-energy terms, since no accurate experimental or theoretical information on the relevant ultracold collisions is presently available. The Raman light fields are represented by the Rabi frequencies \( \Omega_{L1} = 1/2\mu_1 e_{L1} \) and \( \Omega_{L2} = 1/2\mu_2 e_{L2} \), with \( \mu_i \) the two transition dipole matrix elements. Our aim is to find pulses \( e_{L1} \) and \( e_{L2} \) that take the initial population in \( \rho_a \) (atomic BEC) completely over to \( \rho_{13} \) (stable molecular BEC) without populating the intermediate decaying excited state. The starting point is the formulation of a functional that is to be varied in order to obtain the coupled equations solved iteratively in the electric fields:

\[ J = \langle 3 | \rho(t_f) | 3 \rangle - \alpha_1 \int_{t_i}^{t_f} e_{L1}^2(t) dt - \alpha_2 \int_{t_i}^{t_f} e_{L2}^2(t) dt - 2 \text{Re} \left[ \int_{t_i}^{t_f} \text{tr} \{ \dot{\rho}(t) \} \right] + W(\rho(t), t) | \lambda(t) \rangle dt. \]

Here \( \rho(t) \) is a \( 4 \times 4 \) matrix describing the current state, including coherences, of the mixed condensate system governed by Eq. (1). The first term describes the aim, which is to maximize the overlap with the bound molecular state at final time \( t_f \). The next two terms are used to regulate the maximal pulse energy by adequately choosing the parameters \( \alpha_1, \alpha_2 \). The optimization interval is \([t_i, t_f]\) and in the calculations \( t_i = -400 \mu s \) and \( t_f = 100 \mu s \) were chosen.

The functional becomes unconstrained due to the last term, which takes into account that the evolution of \( \rho \) is governed by Eq. (1), written in the form \( \rho(t) = W(\rho(t), t) \). Here \( W(\rho(t), t) \) is the right hand side of Eq. (1) and includes all nonlinear terms. In order to express this constraint to be fulfilled at every time step, a Lagrange multiplier \( \lambda(t) \) is necessary.

Variations with respect to \( \rho, \lambda, \epsilon_{L1}, \) and \( \epsilon_{L2} \) have to be calculated to find the fields that maximize the functional. Beginning with the electric fields the following two equations are obtained:

\[ \Delta \epsilon_{L1}(t) = -\frac{\mu_1}{\alpha_1} \text{Im} \{ \rho_{12}(\lambda_{22} - \lambda_{11} + \lambda_{12}(\rho_{11} - \rho_{22}) - \lambda_{13} \rho_{23}^* - \lambda_{23} \rho_{13}^* + \lambda_{a1} \rho_{a2} + \lambda_{a2} \rho_{a1}) \}, \]
\[ \Delta \epsilon_{L2}(t) = -\frac{\mu_2}{\alpha_2} \text{Im} \{ \rho_{23}(\lambda_{33} - \lambda_{22}) + \lambda_{23}(\rho_{22} - \rho_{33}) + \lambda_{12} \rho_{13}^* + \lambda_{13} \rho_{12}^* + \lambda_{a2} \rho_{a3} + \lambda_{a3} \rho_{a2}^* \}. \]

Clearly these equations, which are used to predict the optimal fields in each iteration, depend on the populations and coherences of the system. In comparison, we also derived the optimal control equations for the GP equations of Ref. [1]:

\[ \Delta \epsilon_{L1}(t) = -\frac{\mu_1}{\alpha_1} \text{Im} \{ \langle \lambda_1 | \phi_2 \rangle + \langle \lambda_2 | \phi_1 \rangle \}, \]
\[ \Delta \epsilon_{L2}(t) = -\frac{\mu_2}{\alpha_2} \text{Im} \{ \langle \lambda_2 | \phi_3 \rangle + \langle \lambda_3 | \phi_2 \rangle \}. \]

Obviously these equations lack the coherence terms. We compared the iteration behavior using Eqs. (4) and (5) with that using Eqs. (6) and (7). The density matrix formalism outperformed the mere population dynamics, since higher yield optimal pulses were obtained with a smaller number of iterations (60 vs thousands). Therefore, not only is the density matrix formulation of the GP equations a physically more complete picture, but the coherence terms lead to optimal control equations especially suited to optimizing STIRAP light fields.

In practice two different ways of iterating on the laser electric fields can be employed. One is the method that implements Eqs. (4) and (5) for the electric fields directly, without memory: \( \epsilon^{(n+1)} = \Delta \epsilon^{(n)} \). It resulted always in a zero light field solution when the decay from the excited molecular state \( \gamma_{sp} \) was turned on. The Krotov method, which was used in further studies, uses instead the electric field equations as correction equations to the field from the last iteration \( \epsilon^{(n+1)} = \epsilon^{(n)} + \Delta \epsilon^{(n)} \). Here \( \epsilon^{(n)} \) stands for the field \( \epsilon_{L1}(t) \) of the \( n \)th iteration and \( \epsilon^{(n+1)} \) for the same fields at the next iteration \( n + 1 \).
Variation with respect to $\lambda$ leads to Eq. (1) with the boundary condition at initial time $\rho(t_f) = \rho_a$. $\rho$ variation leads to the following equations of motion for $\lambda$:

\[
\begin{align*}
    i\dot{\lambda}_a &= -2U_0(\rho_a^{*}\lambda_{a1} + \rho_{a2}\lambda_{a2} + \rho_{a3}\lambda_{a3}) + 2\alpha[\rho_a - 2\rho_1]\lambda_{a1} - 2\rho_{12}\lambda_{a2} - 2\rho_{13}\lambda_{a3} - \text{c.c.,} \\
    i\dot{\lambda}_{11} &= E_{1}^{*}\lambda_{11} - 4\alpha\rho_{a}\lambda_{a1} + \Omega_{L1}\lambda_{12} - \text{c.c.,} \\
    i\dot{\lambda}_{22} &= E_{2}^{*}\lambda_{22} - \Omega_{L1}\lambda_{12} + \Omega_{L2}\lambda_{23} - \text{c.c.,} \\
    i\dot{\lambda}_{33} &= E_{3}^{*}\lambda_{33} - \Omega_{L2}\lambda_{23} - \text{c.c.,} \\
    i\dot{\lambda}_a &= (E_{1}^{*} - 2U_0\rho_a)\lambda_{a1} - \alpha(\lambda_{11} - 2\lambda_{a1}) + \Omega_{L1}\lambda_{a2}, \\
    i\dot{\lambda}_{a2} &= (E_{2}^{*} - 2U_0\rho_a)\lambda_{a2} - \alpha\lambda_{12} + \Omega_{L1}\lambda_{a1} + \Omega_{L2}\lambda_{a3}, \\
    i\dot{\lambda}_{a3} &= (E_{3}^{*} - 2U_0\rho_a)\lambda_{a3} - \alpha\lambda_{13} + \Omega_{L2}\lambda_{a2}, \\
    i\dot{\lambda}_{12} &= (E_{1}^{*} - E_{1})\lambda_{12} - 4\alpha\rho_{a}\lambda_{a2} + \Omega_{L1}(\lambda_{11} - \lambda_{22}) + \Omega_{L2}\rho_{13}, \\
    i\dot{\lambda}_{13} &= (E_{2}^{*} - E_{1})\lambda_{13} - 4\alpha\rho_{a}\lambda_{a3} - \Omega_{L1}\lambda_{12} + \Omega_{L2}\lambda_{22} - \Omega_{L3}\lambda_{33}, \\
    i\dot{\lambda}_{23} &= (E_{3}^{*} - E_{2})\lambda_{23} - \Omega_{L1}\lambda_{13} + \Omega_{L2}\lambda_{22} - \Omega_{L3}\lambda_{33}.
\end{align*}
\]

Due to the nonlinear nature of Eq. (1) the evolution of the Lagrange multiplier $\lambda$ depends on $\rho$ itself and is not independent as for the linear Schrödinger equation. The Lagrange multiplier has to satisfy the boundary condition $\lambda_{i,j}(t_f) = \delta_{i,j}$ at the final time.

The obtained system of four equations [Eqs. (1), (4), (5), and (8)], where Eqs. (1) and (8) depend on the fields $\epsilon_{1}(t), \epsilon_{2}(t)$ given by Eqs. (4) and (5), respectively, is solved as usual by iteration on the electric fields [7]. One iteration is composed of the following steps. Starting with an initial guess pulse, $\rho(t)$ is propagated from its initial value until the final time is reached. These values of $\rho(t)$ at each point of time are stored. Then instead of $\rho(t_f)$ the matrix $\delta_{4,4}$ is used as the boundary condition for $\lambda$ at the final time $t_f$. Both electric fields at $t = t_f$ are calculated using Eqs. (4) and (5) in terms of the Krotov method. These field values are used in conjunction with the stored values of $\rho$ to calculate $\lambda$ backward until the initial time is reached. The improved field obtained is used as the initial guess in the next iteration.

The proposed iteration scheme, one out of three possible in the case of the linear Schrödinger equation, is the only one that works here, since $\lambda$ can only be propagated if $\rho$ is already known, and moreover the propagation will only be well behaved if Eq. (8) is propagated backward in time, due to the $+$ sign of the $\gamma_0$ and $\gamma_p$ decay terms. Numerically the propagation is performed using a variable-order, variable-step Adams method. Iteration was stopped when the yield no longer increased monotonically. Critical to the iteration performance are the values of $\alpha_1$ and $\alpha_2$, which were chosen as close as possible to their thresholds, below which convergence breaks down and strong oscillatory behavior sets in. Too high values will slow down convergence drastically. For the problem at hand $\alpha_1 = \alpha_2 = 2 \times 10^3$ proved to be the best and closest to threshold.

The STIRAP sequence of seven consecutive equidistant pulses of Ref. [1] (gray line in Figs. 1 and 2) transfers 25% of the population into the molecular BEC if a negative magnetic field sweep is applied and 16% in conjunction with a positive sweep. Taking this STIRAP sequence as the initial guess Raman pulse in the optimal control formulation, the yield could be improved upon considerably. The resulting optimal light fields are shown in Figs. 1 and 2, for the case of positive and negative magnetic field sweep, respectively. The optimization was not constrained to amplitudes and pulse separations, but this is mainly the difference between the optimal Raman pulse and the hand-optimized Raman pulse from Ref. [1]. Their structure no longer exists throughout a STIRAP sequence where the second Raman pulse $\epsilon_{L,1}$ precedes the first pulse $\epsilon_{L,1}$. Instead the subpulses have a vary-
FIG. 3. Dynamics of population (a) and coherences (b) induced by optimal pulse for positive magnetic field sweep. Populations: atomic BEC \( \rho_0 \) (thick solid line), quasibound \( \rho_{11} \) (thick dotted line), excited \( \rho_{22} \) (thin solid line), and ground \( \rho_{13} \) (thick dashed line). Coherences: \( \rho_{01} \) (thick solid line), \( \rho_{02} \) (thin solid line), \( \rho_{03} \) (thick dotted line), \( \rho_{12} \) (thin dotted line), \( \rho_{13} \) (thick dashed line), and \( \rho_{23} \) (thin dashed line).

FIG. 4. As Fig. 3 for negative magnetic field sweep.

It should be noted here that optimizing a STIRAP [9] sequence for the \( \Lambda \) system alone is not sufficient for solving the conversion of a BEC to a MBEC efficiently. We verified this, and the light fields for the whole problem differ significantly as they are suitably modulated to adapt to the nonlinearities and the refilling by the atomic BEC in this process.

In conclusion, we have formulated the Gross-Pitaevskii equation modeling the BEC to MBEC conversion via Raman light fields and a magnetic field sweep, using a density matrix formalism. We showed that this is an ideal ansatz for the formulation of the optimal control framework in the case of dissipation. With it we obtained two Raman pulse sequences, each for a different sign of the magnetic field sweep, that both achieve 42% molecular BEC. Therefore we could show that optimal control theory can be derived and applied successfully in the regime of nonlinear Schrödinger equations. STIRAP-like sequences are natural solutions of this density matrix based optimal control formulation and it is not necessary to resort to other proposed, less efficient optimal control schemes.

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