Comment on “Stimulated Raman adiabatic passage from an atomic to a molecular Bose-Einstein condensate”

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(Dated: March 31, 2022)

Collective two-color photoassociation of a freely-interacting ⁸⁷Rb Bose-Einstein condensate is theoretically examined, focusing on stimulated Raman adiabatic passage (STIRAP) from an atomic to a stable molecular condensate. In particular, Drummond et al. [Phys. Rev. A 65, 063619 (2002)] have predicted that particle-particle interactions can limit the efficiency of collective atom-molecule STIRAP, and that optimizing the laser parameters can partially overcome this limitation. We suggest that the molecular conversion efficiency can be further improved by treating the initial condensate density as an optimization parameter.

PACS numbers: 03.75.Nt,03.65.Ge,05.30.Jp,32.80.Wr

Stimulated Raman adiabatic passage (STIRAP) from an atomic to a molecular condensate in the presence of particle-particle interactions was recently investigated. In particular, Drummond et al. predict that, for a ⁸⁷Rb Bose-Einstein condensate (BEC) of typical density ( Prophet Imaging cm⁻³), the STIRAP conversion efficiency is limited by practice in two-photon dephasing caused by particle interactions, and that said limitation can be partially overcome by optimizing the laser parameters. The purpose of this Comment is to suggest that the role of particle-particle interactions can be further downplayed, and the conversion efficiency improved, by treating the initial condensate density as an additional optimization parameter.

The mean-field equations for collective two-color photoassociation of a freely-interacting gas can be written

\[ i \dot{a} = \left( \frac{\Delta}{2} + \Lambda_{aa}|a|^2 + \Lambda_{ag}|g|^2 \right) a - \chi a^* b, \]  
\[ i \dot{b} = \left( \delta - \frac{1}{2} \chi \right) b - \frac{1}{2} \left( \chi aa + \Omega g \right), \]  
\[ i \dot{g} = \left( \Lambda_{ag}|a|^2 + \Lambda_{gg}|g|^2 \right) g - \frac{i}{2} \Omega b, \]

where the complex amplitudes \( a \), \( b \), and \( g \) represent the respective atomic, excited-molecular, and stable-molecular condensates. The laser-matter interactions that drive the respective atom-molecule and molecule-molecule transitions are \( \chi(t) = \chi_0 \exp(-t/T^2) \) and \( \Omega(t) = \Omega_0 \exp(-t/D_2^2/T^2) \), where \( \chi_0 \) includes the effects of Bose-enhancement, i.e., \( \chi_0 \propto \sqrt{\rho} \). The two-photon (intermediate) detuning is \( \Delta \) (\( \delta \)), and the mean-field shift due to atom-atom (atom-molecule, molecule-molecule) interactions is \( \Lambda_{aa} = \rho \lambda_{aa} = 4\pi \hbar \rho a_{aa}/m \) (\( \Lambda_{ag} = \rho \lambda_{ag} = 3\pi \hbar \rho a_{ag}/m \), \( \Lambda_{gg} = \rho \lambda_{gg} = 2\pi \hbar \rho a_{gg}/m \)), where \( m \) is the atom mass and \( a_{aa} \) (\( a_{ag} \), \( a_{gg} \)) is the atom-atom (atom-molecule, molecule-molecule) scattering length. Spontaneous decay is included with the rate \( \gamma_a \), which is generally large enough to justify neglect of any mean-field shifts for the excited-molecular state.

Explicit numbers for ⁸⁷Rb are \( \gamma_s = 7.4 \times 10^7 s^{-1}, \chi_0 = 2.1 \times 10^6 \sqrt{\rho/\rho_0} s^{-1}, \rho_0 = 4.3 \times 10^{14} \text{cm}^{-3}, \lambda_{aa} = 4.96 \times 10^{-11} \text{cm}^{-3}/s, \lambda_{ag} = -6.44 \times 10^{-11} \text{cm}^{-3}/s; \) although unknown, the stable-molecule mean-field shift \( \lambda_{gg} = 2.48 \times 10^{-11} \text{cm}^{-3}/s \) is estimated by assuming equal atom-atom and molecule-molecule scattering lengths.

The idea of optimizing the density of the initial atomic Bose-Einstein condensate is drawn from investigations into forming a molecular condensate via Feshbach-resonant interactions, i.e., magnetoassociation, where collision-induced vibrational relaxation of the molecules is included as a complex particle-particle scattering length, and where a moderate density helps to alleviate the associated irreversible losses.

Consider for example the density \( \rho = 4.3 \times 10^{12} \text{cm}^{-3} \), so that \( \chi_0 = 2.1 \times 10^9 s^{-1}, \lambda_{aa} = 213 s^{-1}, \lambda_{ag} = 107 s^{-1}, \lambda_{gg} = 277 s^{-1}. \) The mean-field shifts are then roughly three orders of magnitude smaller than the peak Bose-enhanced free-bound coupling \( \chi_0 \), and since \( \chi_0 \) sets the timescale for atom-molecule STIRAP, we expect a smaller role for particle interactions compared to when \( \rho = \rho_0 \). This intuition is confirmed in Fig. 1. Note that efficient short-pulse conversion requires asymmetric pulses. The optimal density is \( \rho \sim 10^{12} \text{cm}^{-3} \), for which conversion to a molecular BEC occurs on a timescale \( T = 5 \times 10^3/\chi_0 \sim 50 \text{ms} \). Optimizing the laser parameters should give even further improvement.

Granted, experiments are inhomogeneous and our theory is not. But inhomogeneity may be approximated by averaging over a Thomas-Fermi density distribution, and the subsequent optimal density should be in the same ballpark as the homogeneous prediction. For \( N = 5 \times 10^5 \) atoms in a spherically symmetric trap, reducing the trap frequency from \( \omega_r/2\pi = 100 \text{Hz} \) to \( \omega_r/2\pi \sim 1 \text{Hz} \) decreases the peak density to roughly the optimal value. If the photoassociation laser fails to overlap with the expanded BEC for experimentally reasonable intensities,
The cloudsize could be adjusted by lowering the atom number, or Feshbach-tuning the scattering length \( \tilde{a} \), or a combination thereof.

One-body correlations are neglected as per our original work \(^{[1]}\). Similarly, rogue pair correlations due to photodissociation to noncondensate modes can be prominent for low density \( \rho \); however, the density dependence is weak, and we show elsewhere \(^{[5]}\) that, as expected, rogue correlations are safely neglected for low excited-state fractions \( \rho \ll 0 \). Note that, since a molecule is born of two atoms, \( |g|^2 = 1/2 \) is actually complete conversion. Mean-field shifts are marginalized as the density is decreased, but the timescale for STIRAP \( \propto 1/\sqrt{\rho} \) also increases, and spontaneous decay eventually spoils the show.

FIG. 1: Stimulated Raman adiabatic passage in photoassociation of a freely interacting \(^{87}\)Rb Bose-Einstein condensate. For a given \( \chi_0(\rho) \), the pulse parameters are \( \Omega_0 = 50\chi_0 \), \( T = 5 \times 10^3/\chi_0 \), \( D_1 = 4.5T \) and \( D_2 = 2.5T \). The two-photon (intermediate) detuning is \( \Delta = 0 \) \((\delta = \chi_0)\). Note that, since a molecule is born of two atoms, \( |g|^2 = 1/2 \) is actually complete conversion. Mean-field shifts are marginalized as the density is decreased, but the timescale for STIRAP \( \propto 1/\sqrt{\rho} \) also increases, and spontaneous decay eventually spoils the show.

In conclusion, we suggest that the role of particle-particle collisions can be further downplayed in STIRAP from an atomic to a molecular condensate, and the conversion efficiency thereby improved, by optimizing the initial BEC density along with the laser parameters. Moreover, we have shown elsewhere that Feshbach-tuning the scattering length can improve the molecular conversion efficiency even further \(^{[6]}\). Hence, if all available knobs are tweaked, experiments may yet wind up close to the ideal \(^{[1]}\) of near-unit efficiency.

Support: Academy of Finland (MM, projects 43336 and 50314); Magnus Ehrnrooth Foundation (AC); NSF and NASA (JJ, PHY-0097974 and NAG8-1428); express thanks (from JJ) to Helsinki University of Technology and Matti Kaivola for support and hospitality.

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