

STIMULATED RAMAN ATOM-MOLECULE CONVERSION IN A BOSE-EINSTEIN CONDENSATE

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In the frame of mean field approximation the dynamics of the atoms, molecules and photons in the process of stimulated atom-molecule conversion in the Bose-Einstein condensate was investigated. It was shown that the conversion is both periodic and aperiodic in time. The dynamic rates depend on the initial densities of particles and initial phase difference.

1. Introduction

Experimental observation of the phenomenon of Bose-Einstein condensation (BEC) in dilute, trapped alkali-metal vapors has opened a new context for studying of the properties of mesoscopic systems in the field of low-temperature physics. A fascinating possibility is the observation of new quantum phenomena on the macroscopic scales. It was shown recently that the condensate in double traps can exhibit a quantum coherent atomic oscillatory tunneling between the two traps, if the condensate is initially localized in one well. Due to the nonlinearity these oscillations are suppressed when the density of atoms in the condensate exceeds a critical value. Due to atomic interactions the BEC may exhibit nonlinear wave behavior similar to the self-phase modulation in the field of nonlinear optics.

Below we propose a general type of nonlinearity, which can arise through atom-molecule coupling. This coupling may result in the formation of a molecular Bose condensate through stimulated emission of molecular bosons. Investigation of the dynamics of coupled atom-molecule conversions under the conditions of the Feshbach resonance in magnetic field or in the conditions of atom-molecule resonant optical conversion, accompanied by two Raman electromagnetic pulses, is of special interest. A series of recent experiments on the creation of molecular condensate rely on the application of Feshbach resonance. This coupling mechanism, however, is restricted to the creation of molecules in the highest vibrational levels and is only practicable for a limited number of systems. A more general method is the stimulated optical Raman transition, which can directly produce deeply bound molecules. This method was proposed as a promising way for a fast and efficient conversion of the atomic BEC into a condensate of molecules [1-8]. Such process can give rise to fundamentally new chemistry, so-called “superchemistry”, in which a strong coherent stimulation of chemical reactions is possible via macroscopic occupation of a quantum state by the bosonic chemical species. In other words, superchemistry results on greatly enhanced, non-Arrhenius chemical kinetics at ultralow temperatures. The stimulated Raman atom-molecule conversion can become the dominating mechanism of nonlinearity, which can lead to new chemical reactions. A number of experimental studies of the coherent atom-molecule conversion have shown that direct conversion via Raman photoassociation appears feasible, based on stimulated free-bound and bound-bound transitions in the presence of two laser fields of different

frequencies. Until now it was succeeded to obtain a number of new molecules: K_2 , Na_2 , Li_2 , Cs_2 , Cs_3 , Cs_4 , Rb_2 and He_2 .

2. Main equations

We investigated the dynamics of atom-molecule BEC treating the Raman process of binding of two atoms into molecule as an indivisible process. Here pairs of coherent atoms from the two-atom continuum of the ground electronic potential are transferred: via an excited bound molecular state to a bound molecular state of a lower energy in the ground potential. Raman photoassociation allows coupling to a single molecular state, which can be selected by the Raman laser frequencies. We will show that the initial densities of atoms, molecules, and photons strongly affect the process rate. The reactions may be both periodic and aperiodic in time. Moreover, significant role of the phase difference between the initial components of a reaction is revealed. The macroscopic coherence of the condensate of atoms, molecules, and photons of both pulses predetermine the quantum interference between all components of the reactions and, correspondingly, the importance of phase relations.

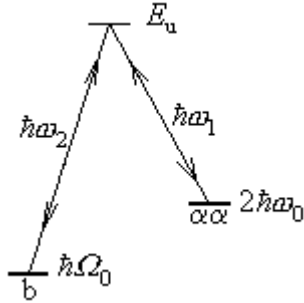


Fig. 1. The scheme of Raman atom-molecule conversion. The $\alpha\alpha$ -level represents the energy level of two free atoms, the b -level is the energy level of molecule.

Let two identical free condensed atoms with zero kinetic energy at zero temperature and total energy $E_i=2\hbar\omega_0$ transfer to a molecular state with the energy $E_m=\hbar\Omega_0$ through a virtual excited molecular state E_u by absorbing and emitting photons with the energies $\hbar\omega_1$ and $\hbar\omega_2$ respectively (Fig. 1). The atom-molecule coupling becomes resonant when the Raman detuning $\delta=\hbar(-2\omega_0+\Omega_0)-\hbar(\omega_2-\omega_1)$ goes to zero. Namely this fact allows to select a single molecular state. We consider the following model interaction Hamiltonian H_{int} , which describes the induced Raman atom-molecule conversion as an indivisible process

$$\hat{H}_{int} = \hbar g(\hat{\alpha}\hat{\alpha}\hat{b}^+\hat{c}_1^+\hat{c}_2^+ + \hat{\alpha}^+\hat{\alpha}^+\hat{b}\hat{c}_1^-\hat{c}_2^-),$$

where $\hat{\alpha}$ and $\hat{b}$ are the boson annihilation operators for atomic and molecular states respectively, $\hat{c}_1$ and $\hat{c}_2$ are the annihilation operators for photons with the frequencies ω_1 and ω_2 , and g is the coupling constant. In essence, we propose the general type of nonlinearity that can lead to the formation of the molecular bosons from the atomic Bose condensate, which is stimulated by two Raman pulses.

We use the mean field approximation, in which the Heisenberg equations of motion for the operators $\hat{\alpha}$, $\hat{b}$, $\hat{c}_1$, and $\hat{c}_2$ are transformed into equations for the amplitudes $\hat{\alpha}$ and $\hat{b}$ of material fields and the amplitudes $\hat{c}_1$ and $\hat{c}_2$ of the electromagnetic fields

$$i\dot{\alpha} = \omega_0\alpha + 2g\alpha^*bc_1^*c_2^*,$$

$$i\dot{b} = \Omega_0b + g\alpha\alpha c_1c_2^*,$$

$$i\dot{c}_1 = \omega_1c_1 + g\alpha^*\alpha^*bc_2^*,$$

$$i\dot{c}_2 = \omega_2c_2 + g\alpha\alpha b^*c_1^*.$$

We supplement this system by the initial conditions

$$\alpha|_{t=0} = \alpha_0 e^{i\varphi_0}, \quad b|_{t=0} = b_0 e^{i\psi_0}, \quad c_1|_{t=0} = c_{10} e^{i\varphi_{10}}, \quad c_2|_{t=0} = c_{20} e^{i\psi_{20}},$$

where each variable is characterized by its initial amplitude and phase. We can see that the stimulated Raman atom-molecule conversion in the condensate is strongly nonlinear in the amplitudes of material and electromagnetic fields. Introducing the particle densities $n=|\alpha|^2$, $N=|b|^2$, $f_1=|c_1|^2$, $f_2=|c_2|^2$ we obtain three integrals of motion $n+2N=n_0+2N_0$, $f_1+N=f_{10}+N_0$ and $f_2-N=f_{20}-N_0$, which are the conservation laws for the particles, and the nonlinear differential equation for the density of molecules

$$\dot{N} = \pm gQ,$$

$$Q^2 = -\left[\frac{\Delta}{g}(N-N_0) - 2n_0 \sqrt{N_0 f_{10} f_{20}} \cos \Theta_0 \right]^2 + 4N(n_0 + 2N_0 - 2N)^2 (N_0 + f_{10} - N)(f_{20} - N_0 + N),$$

where n_0 , N_0 , f_{10} , f_{20} are the initial particle densities, $\theta_0 = \psi_0 - 2\varphi_0 + \psi_{20} - \varphi_{10}$ is the initial phase difference between the material and electromagnetic fields, $\Delta = 2\omega_0 - \Omega_0 + \omega_1 - \omega_2$ is the resonance detuning. Finally we obtain the formal solution in the form of hyperelliptic integral for the molecular density $N(t)$.

3. Discussion of results

Let us analyze the time evolution of the system for the case of exact resonance $\Delta = 0$, where the energy difference between diatomic and molecular states is equal to the energy difference between two photons. We can see that the stimulated Raman atom-molecule conversion is impossible if molecules N and photons f_2 as the products of reaction are simultaneously absent at the initial time. It is due to the complicated inducing of the process. For the beginning of the transformation reaction it is necessary to introduce a certain number of final-state photons f_2 or molecules N into the system. This chemical conversion is dominated by coherent stimulated process, in which transitions are enhanced by the number of molecules or photons f_2 in the product mode. This fact is completely different from the usual chemical kinetics, which predicts that the rates of chemical reactions do not depend on the number of particles in the product mode and go to zero at low temperatures according to the Arrhenius law.

Having no opportunity to calculate the hyperelliptical integral for the density of molecules, further we consider the time evolution for the different relations between the initial densities of particles. Let us consider the time evolution in the approximation of the given density of photons of both pulses $f_{10}, f_{20} \gg N_0, n_0$. The solution depends on the initial phase difference. In the case when phase difference $\Theta_0 = \pm\pi/2$ solution for the density of molecules has the form

$$N = \left(N_0 + \frac{n_0}{2} \right) \cdot \left[\frac{\sqrt{N_0} \pm \sqrt{N_0 + n_0/2} \cdot \text{th} \left(2g \sqrt{f_{10} f_{20}} (N_0 + n_0/2) t \right)}{\sqrt{N_0 + n_0/2} \pm \sqrt{N_0} \cdot \text{th} \left(2g \sqrt{f_{10} f_{20}} (N_0 + n_0/2) t \right)} \right]^2,$$

where the signs (+) and (-) correspond to the initial conditions $\dot{N}|_{t=0} > 0$ and $\dot{N}|_{t=0} < 0$ or to the difference phases $\Theta_0 = -\pi/2$ and $\Theta_0 = +\pi/2$. The solution with sign (+) for $\Theta_0 = -\pi/2$ increases monotonously in time and approaches the value $N_0 + n_0/2$. All atoms convert into molecules and the process terminates (Fig. 2). The rate of time evolution increases with increasing of densities N_0 , n_0 , f_{10} , and f_{20} . As for the solution with sign (-), we can see that at first

it decreases in time, approaches zero, then increases and asymptotically reaches the same limiting value $N_0 + n_0/2$. The reverse reaction accompanied by the dissociation of molecules does not proceed. The process of atom conversion into molecules has the aperiodic character.

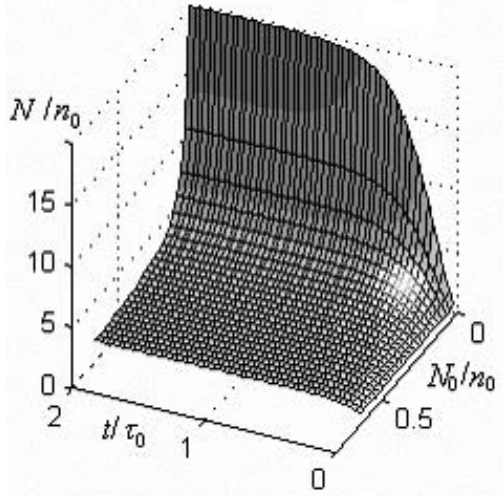


Fig. 2. The dependence of the normalized concentration N/n_0 in approximation $f_{10}, f_{20} \gg N_0, n_0$ on the normalized time t/τ_0 ($\tau_0 = (gn_0^{3/2})^{-1}$) and normalized initial concentration of molecules N_0/n_0 for the values of parameters $\Theta_0 = \pm\pi/2, f_{10}/n_0 = f_{20}/n_0 = 2$.

In the case when the initial phase difference $\Theta_0 = \pm k\pi$ the solution essentially depends on the relation between the densities N_0 and n_0 . It has the periodic character, the amplitude and period of oscillations of which depend on the initial densities of particles. The amplitude of oscillations at first decreases with the increasing of the ratio n_0/N_0 , reaches zero at $n_0/N_0 = 4$ and then increases.

From the obtained results we can see that for the nonzero densities of all particles, when the relation $n_0 = 4N_0$ is fulfilled, the system does not change its values in time. We can affirm that when $n_0 = 4N_0$ and $\Theta_0 = \pm k\pi$ the system is balanced in such a manner that its amplitude of oscillation is found to be zero with nonzero period of oscillation. At the fixed values of N_0, n_0, f_{10} , and f_{20} the change of initial phase difference Θ_0 leads to the changing both the period and the amplitude of oscillations. The density of molecules changes in the limits from $N_{\min}$ till $N_{\max}$, which are determined by the initial densities of particles and phase difference Θ_0 . This fact evidences the possibility of phase control of the time evolution of the system. We can govern the time evolution of the system by the initial phase difference varying.

In general, we observe the oscillations between atomic and molecular condensates, which provide clear evidence of a long-range coherence effect. In contrast, stimulated Raman photoassociation in a thermal cloud of atoms would not produce similar collective oscillations, because the phases associated with the individual atom-molecule conversion processes are random. Stimulated atom-molecule conversion in a condensate is nonlinear process in the atomic and molecular amplitudes. Important signatures of the coherent stimulation are the dependence of the nonlinear period oscillations and the reaction rate on the densities of particles.

We can study the time evolution of the system in the approximation of a given density of only one of applied pulses too. Let us suppose that it is the second pulse and let $f_{20} \gg f_{10}, n_0, N_0$. In this case until $f_{10} < n_0/2$ the density of molecules oscillates in the limits between zero and $N_0 + f_{10}$ (Fig. 3). The period of oscillations depends on the particle densities: $T = \pi / (2g\sqrt{f_{20}(N_0 + n_0/2)(n_0/2 - f_{10})})$. It increases if f_{10} increases and diverges when $f_{10} \rightarrow n_0/2$. When $f_{10} > n_0/2$ the time evolution of the system is found to be aperiodic one. All atoms convert into molecules, the density of molecules monotonously reaches the value

$N_0+n_0/2$ and by this the conversion process finishes. The evolution in the reverse direction accompanied by the dissociation of molecules does not take place.

We point out that in the approximation of the given density of the photons of the first pulse $f_{10} \gg f_{20}$, N_0 , n_0 the time evolution of the system is always aperiodic one, accompanied by the increasing of the density of molecules. The time evolution takes place only in one direction. The formed molecules do not dissociate.

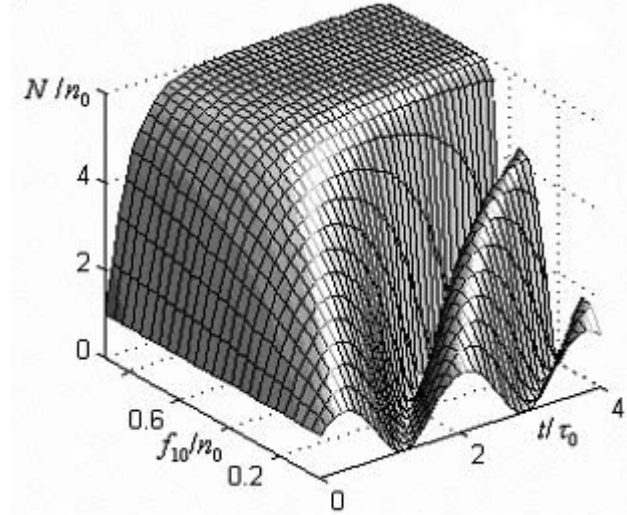


Fig. 3. The dependence of the normalized concentration N/n_0 in approximation $f_{20} \gg f_{10}$, n_0 , N_0 on the normalized time t/τ_0 ($\tau_0 = (gn_0^{3/2})^{-1}$) and on the normalized concentration of photons of the first pulse f_{10}/n_0 for the values of parameters $\Theta_0 = \pi/2$, $f_{20}/n_0=3$.

Let us consider the approximation of the given density of atoms: $n_0 \gg N_0$, f_{10} , f_{20} . The time evolution has an interesting behavior for the initial phase difference $\Theta_0 = \pi/2$. For $N_0 > f_{20}$ the density of molecules oscillates above some background density $N_0 - f_{20}$ and changes between the values $N_0 - f_{20}$ and $N_0 + f_{10}$. When $N_0 < f_{20}$ the molecular background disappears and the density of molecules oscillates between zero and $N_0 + f_{10}$. If $N_0 = f_{20}$ the time evolution has the aperiodic character (Fig. 4). Period of oscillation changes nonmonotonously depending on the ratio f_{20}/N_0 . It first increases with the increase of the ratio f_{20}/N_0 , diverges at $f_{20}/N_0=1$ and then decreases at $f_{20} > N_0$.

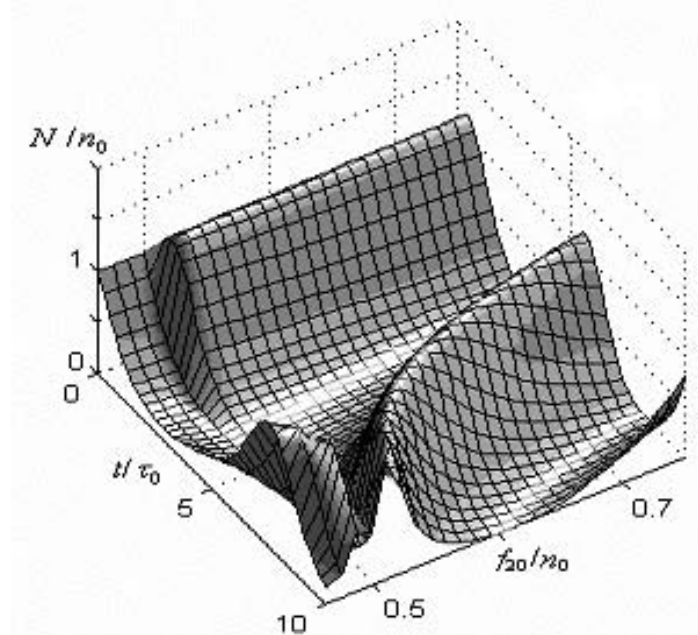


Fig. 4. The dependence of the normalized concentration N/n_0 in approximation $f_{10} \gg f_{20}$, n_0 , N_0 on the normalized time t/τ_0 ($\tau_0 = (gn_0^{3/2})^{-1}$) and on the normalized concentration of photons of the second pulse N/n_0 for the values of parameters $\Theta_0 = \pi/2$, $f_{20}/n_0=3$.

The time evolution for the initial phase difference $\Theta_0 = \pm k\pi$ has the periodic behaviour for arbitrary relations between the particle densities. Both amplitude and period of the oscilla-

tions depend on the initial densities. In this case there also exists the regime of complete stopping of the evolution or the oscillatory regime with zero amplitude, but nonzero period. Changing the initial densities so that the relation $f_{20}=f_{10}+f_{10}f_{20}/N_0$ is fulfilled we can observe that the system conserves the particle density and it does not evolutionize. In the frame of this approximation we also observe the change of regime of oscillations by changing only the initial phase difference Θ_0 .

4. Conclusion

In conclusion we estimate the oscillation frequency in the system of Bose-condensed atoms and molecules $^{87}\text{Rb}_2$. Comparing our results with the results from [1] we determine the value of the coupling constant $g=8\times 10^{-18} \text{ cm}^{9/2}\text{s}^{-1}$. We can obtain the period of oscillation from the simple expression: $T_0 = \pi/g n_0 \sqrt{f_0}$. For the densities $n_0 \approx f_0 \approx 10^{14} \text{ cm}^{-3}$ we obtain $T_0 \approx 4 \times 10^{-4} \text{ s}$ and the frequency of order $2 \times 10^4 \text{ s}^{-1}$.

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