Quantum effects on the dynamics of a two-mode atom-molecule Bose-Einstein condensate

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We study the system of coupled atomic and molecular condensates within the two-mode model and beyond mean-field theory. Large-amplitude atom-molecule coherent oscillations are shown to be damped by the rapid growth of fluctuations near the dynamically unstable molecular mode. This result contradicts earlier predictions about the recovery of atom-molecule oscillations in the two-mode limit. The frequency of the damped oscillation is also shown to scale as $\sqrt{N}/\ln N$ with the total number of atoms N, rather than the expected pure $\sqrt{N}$ scaling. Using a linearized model, we obtain analytical expressions for the initial depletion of the molecular condensate in the vicinity of the instability, and show that the important effect neglected by mean-field theory is an initially nonexponential ''spontaneous'' dissociation into the atomic vacuum. Starting with a small population in the atomic mode, the initial dissociation rate is sensitive to the exact atomic amplitudes, with the fastest (superexponential) rate observed for the entangled state formed by spontaneous dissociation.

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Recent photoassociation [1] and Feshbach resonance [2,3] experiments suggest the possibility of producing molecular Bose-Einstein condensates (BEC's) [4–9]. Large-amplitude coherent oscillations between an atomic BEC and a molecular BEC have been theoretically predicted [4–6]. A common theme to these studies is the use of the Gross-Pitaevskii (GP) mean-field theory (MFT), reducing the full multibody problem into a set of two coupled nonlinear Schrödinger equations. These are then solved numerically to obtain the Josephson-type dynamics of the coupled atomic and molecular fields.

The simple GP dynamics is substantially affected by condensate depletion due to inelastic collisions [5,7,10], spontaneous emission, and the inclusion of noncondensate modes [10–14]. Consequently, the atom-molecule oscillations are expected to be strongly damped under current experimental conditions. The proposed remedy for this detrimental effect involves a recovery of an effective two-mode dynamics [13], thereby preventing the buildup of thermal population.

In this article we point out that even in the perfect two-mode limit, MFT fails to provide long-term predictions due to strong interparticle entanglement near the dynamically unstable molecular mode. Quantum corrections to MFT appear in the vicinity of its dynamical instabilities on time scales that grow only logarithmically with the number N of condensate particles [15–17]. Thus, even in the absence of a ''proper'' thermal bath of noncondensate states, the mean-field equations are coupled to a reservoir of Bogoliubov fluctuations [16,18]. The rapid growth of these fluctuations near the instability is analogous to the rapid population of the thermal cloud, similarly inhibiting the mean-field atom-molecule oscillations. Our results, obtained using the numerical solution of exact quantum equations, go beyond the Hartree-Fock-Bogoliubov approach [12]. The leading quantum effect is identified as a nonexponential spontaneous decay of the molecular condensate and the dynamics is shown to be highly sensitive to the initial conditions. We note that similar quantum corrections are well known for parametric oscillations in quantum optics [19–22].

We consider the simplest model of the atom-molecule condensate, in which particles can only populate two second-quantized modes: an atomic mode, associated with the creation and annihilation operators $\hat{a}$ and $\hat{a}^\dagger$ and a molecular mode, associated with the creation and annihilation operators $\hat{b}$ and $\hat{b}^\dagger$. The two modes are coupled by means of a near-resonant two-photon transition or a Feshbach resonance, with a coupling frequency Ω and detuning Δ. Setting the zero energy to the energy of the molecular mode the two-mode Hamiltonian reads

$$\hat{H}=\frac{\hbar\Delta}{2}\hat{a}^\dagger\hat{a}+\frac{\hbar\Omega}{2}(\hat{a}^\dagger\hat{a}^\dagger\hat{b}+\hat{b}^\dagger\hat{a}\hat{a}). \tag{1}$$

For $\Delta=0$, the Hamiltonian of Eq. (1) is identical [4,23] to the well-known Hamiltonian describing the optical process of parametric oscillations [24], where dissociation is equivalent to parametric down-conversion and association is the analog of second-harmonic generation. We will take Ω to be real and positive without loss of generality, since the relative phase between the modes is determined up to an additive constant and the overall sign of H is insignificant.

We obtain a generalization of the Bloch representation for the two-mode system (similarly to the approach taken in Refs. [16,17]) by introducing the three operators

$$\hat{\mathcal{L}}_x\equiv\sqrt{2}\,\frac{\hat{a}^\dagger\hat{a}^\dagger\hat{b}+\hat{b}^\dagger\hat{a}\hat{a}}{N^{3/2}},$$

$$\hat{\mathcal{L}}_y\equiv\sqrt{2}\,\frac{\hat{a}^\dagger\hat{a}^\dagger\hat{b}-\hat{b}^\dagger\hat{a}\hat{a}}{iN^{3/2}},$$

$$\hat{\mathcal{L}}_z\equiv\frac{2\hat{b}^\dagger\hat{b}-\hat{a}^\dagger\hat{a}}{N}, \tag{2}$$

where N denotes the total number of atoms. Discarding c-number terms, the Hamiltonian of Eq. (1) then takes the simple form

$$\hat{H}=\hbar\left[\left(\frac{N}{2}\right)^{3/2}\Omega\hat{\mathcal{L}}_x-\frac{N}{4}\Delta\hat{\mathcal{L}}_z\right], \tag{3}$$

and the Heisenberg equations of motion for the three operators of Eq. (2) read

$$\frac{d}{dt}\hat{\mathcal{L}}_x=-\Delta\hat{\mathcal{L}}_y,$$

$$\frac{d}{dt}\hat{\mathcal{L}}_y=-\frac{3}{4}\sqrt{2N}\Omega\left[(\hat{\mathcal{L}}_z-1)\left(\hat{\mathcal{L}}_z+\frac{1}{3}\right)\right]+\Delta\hat{\mathcal{L}}_x+\sqrt{\frac{2}{N}}\Omega,$$

$$\frac{d}{dt}\hat{\mathcal{L}}_z=-\sqrt{2N}\Omega\hat{\mathcal{L}}_y. \tag{4}$$

These three operators do *not* represent SU(2), but all three commute with the conserved total atom number $N=\hat{a}^\dagger\hat{a}+2\hat{b}^\dagger\hat{b}$.

The mean-field approximation is invoked by approximating second-order expectation values $\langle\hat{\mathcal{L}}_i\hat{\mathcal{L}}_j\rangle$ as products of the first-order moments $\langle\hat{\mathcal{L}}_i\rangle$ and $\langle\hat{\mathcal{L}}_j\rangle$ [16,17],

$$\langle\hat{\mathcal{L}}_i\hat{\mathcal{L}}_j\rangle\approx\langle\hat{\mathcal{L}}_i\rangle\langle\hat{\mathcal{L}}_j\rangle. \tag{5}$$

Approximation (5) is correct to $O(1/\sqrt{N})$. Thus we can also neglect the *c*-number term $\sqrt{2/N}\Omega$ in Eq. (4). Defining $\vec{\mathbf{s}}\equiv(\langle\hat{\mathcal{L}}_x\rangle,\langle\hat{\mathcal{L}}_y\rangle,\langle\hat{\mathcal{L}}_z\rangle)$, rescaling the time as $\tau=\sqrt{N}\Omega t$, and using Eq. (5), we obtain the mean-field equations

$$\frac{d}{d\tau}s_x=-\delta s_y,$$

$$\frac{d}{d\tau}s_y=-\frac{3\sqrt{2}}{4}(s_z-1)\left(s_z+\frac{1}{3}\right)+\delta s_x,$$

$$\frac{d}{d\tau}s_z=-\sqrt{2}s_y, \tag{6}$$

where the dimensionless rescaled detuning δ is defined as $\delta\equiv\Delta/(\sqrt{N}\Omega)$ [Eqs. (6) are equivalent to Eqs. (32) of Ref. [10] without the inelastic-collision terms]. Lyapunov analysis of Eqs. (6) shows that as long as $|\delta|<\sqrt{2}$ the stationary point $\vec{\mathbf{s}}=(0,0,1)$ corresponding to the entire population being in the molecular mode, is (as pointed out for the idler mode of parametric oscillations [20,21]) dynamically unstable because any small perturbation to the mean-field equations (6) would trigger the parametric oscillation. In the vicinity of this point, MFT is expected to break down on a time scale that is only logarithmic in N [15–17]. In order to verify this prediction, we solve the full N-body problem by fixing N, thereby restricting the available phase space to Fock states of the type $|n,(N-n)/2\rangle$ with n atoms and $(N-n)/2$ molecules, where $n=0,2,4,\ldots,N$. Thus we obtain an $(N/2+1)$-dimensional representation for the Hamiltonian and the N-body density operator $\hat{\rho}$. The quantum solution is then obtained by numerically propagating $\hat{\rho}$ according to the Liouville–von Neumann equation

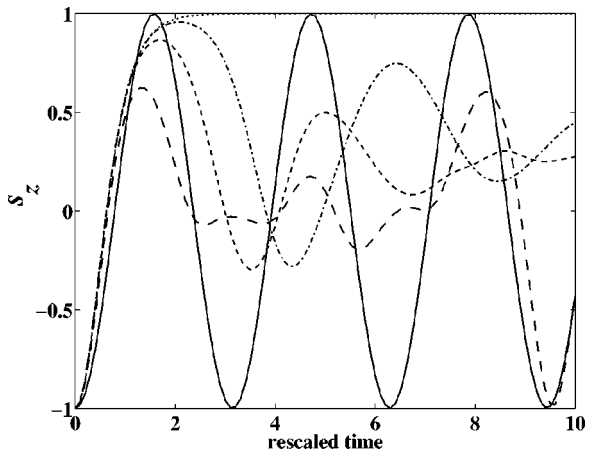

FIG. 1. Numerically calculated population imbalance between atoms and molecules, as a function of the rescaled time τ for 2 (—), 10 (– – –), 100 (- - -), and 1000 (-·-) particles, compared with the mean-field prediction (· · ·).

$$i\hbar\dot{\hat{\rho}}=[\hat{H},\hat{\rho}], \tag{7}$$

and the expectation values of the three operators of Eq. (2) are retrieved as $s_i=\mathrm{Tr}(\rho\hat{\mathcal{L}}_i)$.

In what follows, we shall assume that $\delta=0$, as required to obtain unit-amplitude atom-molecule oscillations. In Fig. 1 we plot the expectation value s_z corresponding to the population difference between the modes as a function of the rescaled time τ for various values of the total particle number. The initial conditions are $\vec{\mathbf{s}}=(0,0,-1)$ corresponding to an initially populated atomic mode. The mean-field solution (dotted line) depicts the convergence of s_z to the unstable fixed point. The quantum solutions (identical to the results of Ref. [4] obtained by solving the N-particle Schrödinger equation) initially follow the mean-field evolution closely. However, in the vicinity of the molecular mode ($s_z=1$) the quantum trajectories break away from the mean-field prediction on a time scale that grows only logarithmically with N. Thus, the oscillation frequency scales with $\sqrt{N}/\ln(N)$ as opposed to the expected scaling with $\sqrt{N}$. Moreover, the oscillations are damped by the strong entanglement near the molecular mode, in complete analogy with the damping of oscillations when the two-mode system is coupled to external thermal modes. Full Rabi-type coherent oscillations can only be observed for a single pair of atoms.

We note that the results of Fig. 1 cannot be reproduced by a Hartree-Fock-Bogoliubov approach [12]. In order to obtain the damping of coherent oscillations, one has to go deeper into the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy of equations of motion, and maintain a number of equations comparable to the total number of particles N.

Equations (4) are equivalent to the equations of motion for the two annihilation operators $\hat{a}$ and $\hat{b}$,

$$i\dot{\hat{a}} = \frac{\Delta}{2}\hat{a} + \Omega \hat{b}\hat{a}^{\dagger}, \tag{8a}$$

$$i\dot{\hat{b}} = \frac{\Omega}{2}\hat{a}\hat{a}. \tag{8b}$$

To appreciate why MFT fails as it does, we will now focus our attention on the vicinity of the dynamically unstable all-molecule state, using a linearized model in which the molecular annihilation operator $\hat{b}$ is replaced by a c number b of $O(\sqrt{N/2})$. In this approximation, which is valid as long as the population of the molecular state is large and the effect of its depletion on the atomic population growth rate can be neglected, Eq. (8a) becomes

$$i\dot{\hat{a}} = \frac{\Delta}{2}\hat{a} + \Omega b\hat{a}^{\dagger}. \tag{9}$$

Equation (9) in combination with its complex conjugate provides an autonomic set of two linear operator equations, which can be solved using common methods reducing it to an eigenproblem. When the molecular mode is dynamically unstable ($|\delta| < \sqrt{2}$) we have

$$\Omega|b| > |\Delta|/2, \tag{10}$$

and the exact solution of Eq. (9) takes the rapidly growing form

$$\hat{a}(t) = \hat{a}(0)\cosh\lambda t - \frac{i}{\lambda}\left[\frac{\Delta}{2}\hat{a}(0) + \Omega b\hat{a}^{\dagger}(0)\right]\sinh\lambda t, \tag{11}$$

where

$$\lambda = \sqrt{\Omega^2|b|^2 - (\Delta/2)^2}. \tag{12}$$

The time dependence of the atom number operator $\hat{n}_a(t) = \hat{a}^{\dagger}(t)\hat{a}(t)$ is thus given as

$$\langle \hat{n}_a(t)\rangle = n_{\rm sp}(t) + n_{\rm st}(t), \tag{13}$$

where the term

$$n_{\rm sp}(t) = \frac{\Omega^2|b|^2}{\lambda^2}\sinh^2\ \lambda t, \tag{14}$$

not accounted for by MFT, corresponds to spontaneous dissociation into the atomic vacuum and the term

$$n_{\rm st}(t) = \langle \hat{n}_a(0)\rangle\left(\cosh 2\lambda t + \frac{\Delta^2}{2\lambda^2}\sinh^2\lambda t\right) - \frac{\Omega}{\lambda}{\rm Im}\left[\langle \hat{a}(0)\hat{a}(0)\rangle b^*\left(\sinh 2\lambda t - i\frac{\Delta}{\lambda}\sinh^2\lambda t\right)\right] \tag{15}$$

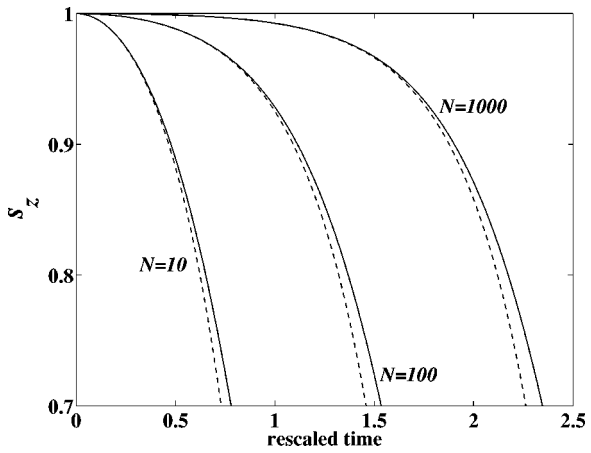

FIG. 2. Depletion of the purely molecular mode for $N=10$, 100, and 1000 particles, according to Eq. (16) (- - -), compared with "exact" numerical results (—).

depicts stimulated dissociation taking place when the atomic state is initially populated. The two terms on the right-hand side of Eq. (15) correspond to noncoherent and coherent initial occupation, respectively.

Starting from the dynamical instability ($|b| = \sqrt{N/2}$) with zero atomic occupation, and assuming zero detuning, the initial evolution of the expectation value $s_z(t)$ is given according to Eqs. (13)–(15) by the purely spontaneous process,

$$s_z(t) = 1 - \frac{2}{N}\sinh^2(\tau/\sqrt{2}). \tag{16}$$

According to Eq. (16) the initial decay of the atomic mode is nonexponential as the leading decay term is quadratic rather than linear in t. This behavior is in accordance with the initial nonexponential decay of a general spontaneous-emission process [25]. The quadratic gain of the atomic population is confirmed by the results of Fig. 2, where we compare the initial depletion of the molecular mode according to Eq. (16) with exact quantum results obtained for various values of N. The agreement is initially excellent until the occupation of the atomic mode becomes significant compared to the molecular occupation. Moreover, it is evident from Eq. (16) and confirmed by the results of Fig. 2, that the time at which the quantum spontaneous-emission term will become significant, grows only logarithmically with N, in agreement with our prediction.

Equation (14) was already obtained in Ref. [13], as the asymptotic expression at $t \to \infty$. However, at this limit, the atomic occupation becomes comparable to the molecular population, and the depletion of the molecular mode should be taken into account. Nevertheless, Eq. (14) is an exact solution to the model of Eq. (9), applicable at small t.

It is interesting to note that when $|\delta| > \sqrt{2}$ the molecular mode is stabilized. Consequently, the exact solution of Eq. (9), when

$$\Omega|b| < |\Delta|/2, \tag{17}$$

is an oscillatory function of the form

$$\hat{a}(t)=\hat{a}(0)\cos|\lambda|t-\frac{i}{|\lambda|}\left[\frac{\Delta}{2}\hat{a}(0)+\Omega b\hat{a}^{\dagger}(0)\right]\sin|\lambda|t, \tag{18}$$

where now

$$|\lambda|=\sqrt{(\Delta/2)^2-\Omega^2|b|^2}. \tag{19}$$

The watershed value of $|\delta|=\sqrt{2}$ agrees with previous results obtained by analysis of the Bogoliubov spectrum of photoassociation [23]. It is also worth noting that as shown in Fig. 1, the evolution of a single molecule coupled to a single atomic mode is always described by an oscillating solution, similar to Eq. (18) since Bose enhancement, depicted by the exponential gain of Eqs. (11) and (13) is a collective effect, analogous to lasing.

Finally, we consider the dependence of the stimulated-decay term, as described by Eq. (15), on the exact initial conditions. Starting with an initially small coherent atomic amplitude $[|\langle\hat{a}(0)\hat{a}(0)\rangle|=\langle\hat{n}_a(0)\rangle]$ and assuming exact resonance ($\Delta=0$), the initial evolution of $n_{\rm st}$ can be varied from exponential gain to exponential decay $[n_{\rm st}(t)\sim\exp(\pm\lambda t)]$ by controlling the relative phase between the atomic and molecular modes $[\langle\hat{a}(0)\hat{a}(0)\rangle$ and $b=\langle\hat{b}\rangle]$. Moreover, the squeezed state, formed by spontaneous dissociation, always gains faster $[\dot{n}_{\rm sp}(t)/n_{\rm sp}(t)=2\lambda\coth\lambda t>2\lambda]$, due to its high degree of entanglement $[|\langle\hat{a}(t)\hat{a}(t)\rangle|/\langle\hat{n}_a(t)\rangle=\coth\lambda t>1]$. In Fig. 3 we compare the predictions of Eq. (13) with exact quantum results for three initial amplitudes, corresponding to the same initial atomic occupation of $4/3N$. The (a) and (b) curves correspond to states of the form $c_0|0,N/2\rangle+c_2|2,N/2-1\rangle$ with $(c_0=\sqrt{2/3},c_2=i\sqrt{1/3})$ and $(c_0=\sqrt{2/3},c_2=-i\sqrt{1/3})$, respectively, giving $\hat{b}^*\langle\hat{a}(0)\hat{a}(0)\rangle=\pm i|b|\langle\hat{n}_a(0)\rangle$ (in equivalence to coherent states). The (c) lines depict the continued decay of a squeezed state with the same $\langle\hat{n}_a(0)\rangle$, formed by the purely spontaneous process, starting at $\tau=-0.53$. As expected, an initial exponential atomic loss is observed when $c_2=i\sqrt{1/3}$ and an exponential atomic gain is observed when $c_2=-i\sqrt{1/3}$. The gain of the spontaneously produced squeezed state, depicted by Eq. (14), is as we pointed out before—initially nonexponential, becoming so only at later times.

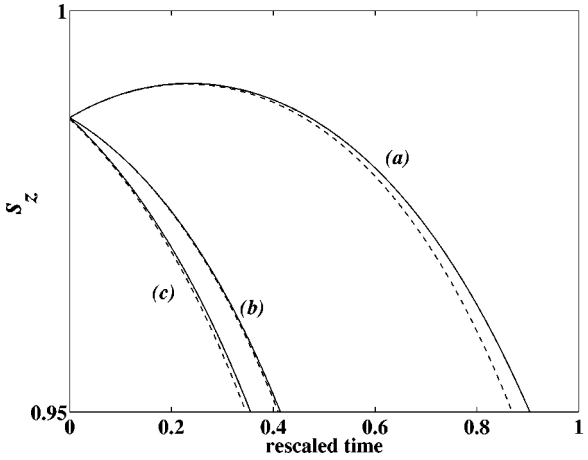

FIG. 3. Depletion of the molecular mode for three 100-particle states with the same initial value of $s_z(0)=74/75$: (a) $(2/3)^{1/2}|0,N/2\rangle+i(1/3)^{1/2}|2,N/2-1\rangle$, (b) $(2/3)^{1/2}|0,N/2\rangle-i(1/3)^{1/2}|2,N/2-1\rangle$, and (c) the state created by the spontaneous process starting from the pure molecular mode, according to Eq. (15) (- - -). Corresponding solid lines are "exact" numerical results.

In conclusion, due to the dynamical instability of the molecular mode, the atomic ensemble produced by the dissociation stage of the parametric oscillation is highly entangled. Consequently, atom-molecule coherent oscillations are predicted to be damped even in the two-mode limit. The leading quantum correction is an initially nonexponential spontaneous decay, becoming significant on a timescale that only grows as $\ln N$. Stimulated processes taking place when there is an initial atomic population are sensitive to different initial conditions corresponding to the same $s_z(0)$. The fastest (superexponential) decay rate is obtained for the initial state formed by spontaneous dissociation. This dependence originates in the parametric-oscillation phase, driving it towards the molecular mode or away from it. It has significant implications on the case of multiple atomic modes, suggesting that the rapid growth observed when these modes are initially partially populated [13] would be sensitive to the exact atomic amplitudes.

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[1] R. Wynar *et al.*, Science **287**, 1016 (2000).
[2] S. Inouye *et al.*, Nature (London) **392**, 151 (1998); J. Stenger *et al.*, Phys. Rev. Lett. **82**, 4569 (1999).
[3] S.L. Cornish *et al.*, Phys. Rev. Lett. **85**, 1795 (2000).
[4] J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999).
[5] E. Timmermans *et al.*, Phys. Rev. Lett. **83**, 2691 (1999); Phys. Rep. **315**, 199 (1999).
[6] D.J. Heinzen *et al.*, Phys. Rev. Lett. **84**, 5029 (2000).
[7] V.A. Yurovsky *et al.*, Phys. Rev. A **60**, R765 (1999).
[8] F.A. van Abeelen and B.J. Verhaar, Phys. Rev. Lett. **83**, 1550 (1999).
[9] C.J. Williams and P.S. Julienne, Science **287**, 986 (2000).
[10] V.A. Yurovsky *et al.*, Phys. Rev. A **62**, 043605 (2000).
[11] F.H. Mies, P.S. Julienne, and E. Tiesinga, Phys. Rev. A **61**, 022721 (2000).
[12] M. Holland, J. Park, and R. Walser, Phys. Rev. Lett. **86**, 1915 (2001).
[13] K. Góral, M. Gajda, and K. Rzażewski, Phys. Rev. Lett. **86**, 1397 (2001).
[14] J.J. Hope and M.K. Olsen, Phys. Rev. Lett. **86**, 3220 (2001).

[15] Y. Castin and R. Dum, Phys. Rev. Lett. **79**, 3553 (1997).
[16] A. Vardi and J.R. Anglin, Phys. Rev. Lett. **86**, 568 (2001).
[17] J. R. Anglin and A. Vardi, Phys. Rev. A **64**, 013605 (2001).
[18] S. Habib *et al.*, Phys. Rev. Lett. **79**, 6 (1997).
[19] D.F. Walls and R. Barakat, Phys. Rev. A **1**, 446 (1970).
[20] D.F. Walls, Phys. Lett. **32A**, 476 (1970).
[21] D.F. Walls and C.T. Tindle, J. Phys. A **5**, 534 (1972).
[22] M. Fleischhauer and O. Veits, Z. Naturforsch., A: Phys. Sci. **54**, 57 (1999).
[23] M. Kostrun *et al.*, Phys. Rev. A **62**, 063616 (2000).
[24] A. Yariv, *Quantum Electronics* (Wiley, New York, 1989).
[25] A. I. Baz, Ya. B. Zeldovich, and A. M. Perelomov, *Scattering, Reactions and Decay in Nonrelativistic Quantum Mechanics* (Nauka, Moscow, 1971).