Nonlinear dynamical instabilities of a condensate system in an atom-molecule dark state

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(Received 19 October 2007; revised manuscript received 19 February 2008; published 8 April 2008)

We study dynamics of a nonlinear three-level Λ system describing Bose-Einstein condensates of atoms and diatomic molecules coupled by a two-color laser field. The system has a nonlinear dark state which is a generalization of the usual atomic dark state. In the recent paper [A. P. Itin and S. Watanabe, Phys. Rev. Lett. **99**, 223903 (2007)], nonlinear instabilities of the dark state due to 1:1 and 1:2 resonances were discussed in the model without mean-field collisional interactions. Here we investigate the dark state of a model with collisional interactions. We show that nonlinear instabilities can be used, in particular, for precise determination of the scattering lengths.

DOI: 10.1103/PhysRevA.77.043610 PACS number(s): 03.75.Mn, 03.75.Nt, 02.30.Oz

I. INTRODUCTION

In the last decade there has been explosive growth of interest in quantum properties of Bose-Einstein condensates (BECs) and in their mean-field dynamics [1]. The nonlinearity results in a variety of exciting phenomena, among which we mention instabilities of matter waves, bright and dark solitons (see, e.g., [2] for the recent reviews of these issues, and [3–5], respectively, for some particular examples), collapse (see, e.g., the review [6] and references therein), vortex dynamics [7], shock waves [8], etc. However, numerous theoretical studies mostly deal with effectively *linear* dynamics of small excitations of ground states of the respective nonlinear systems (employing, say, Bogolyubov-de-Gennes equations, or other schemes of linearizations). While even the linear dynamics is nontrivial and often provides one with useful information it sometimes appears to be insufficient to reproduce the authentic *nonlinear* dynamics which goes *beyond* the standard linearization.

On the other hand a BEC represents a remarkable object for testing the ideas of the nonlinear science and in particular for theoretical and experimental study of the linear and nonlinear instabilities. In the present work we address one of such systems and present some counterintuitive and intrinsically nonlinear phenomena appearing in atomic-molecular BECs.

More specifically we consider a mixture of atomic, excited molecular, and ground-state molecular condensates coupled by two-color Raman photoassociation. The related three-level Λ system is depicted schematically in Fig. 1(a). The state $|a\rangle$ corresponds to the atomic BEC which is coupled to the excited diatomic molecular condensate (the state $|e\rangle$) via a Raman laser pulse Ω_p ("pump"). Without the BEC (with thermal atoms), the free-bound transition would be very weak. However, since all atoms in $|a\rangle$ are in the same state and transferred to $|e\rangle$ coherently, the efficiency of the process is greatly enhanced (see, e.g., [9–12]). The state $|e\rangle$ is coupled to the ground state of the molecular BEC by a "dump" pulse Ω_d. The system has already received a considerable amount of attention, in particular due to prospects of superchemistry with molecular condensates [13] and precise determination of *s*-wave scattering lengths [14]. Its linear counterpart is widely used in quantum manipulations, in particular in the STIRAP (stimulated Raman adiabatic passage [15]) process.

The STIRAP process in Λ systems is based on the existence of the dark state which is a superposition of only $|a\rangle$ and $|g\rangle$ states. The dark state is determined by instantaneous values of $\Omega_{p,d}$; ideally, population transfer happens via the dark state as $\Omega_{p,d}$ are slowly changed, with the $|e\rangle$ state re-

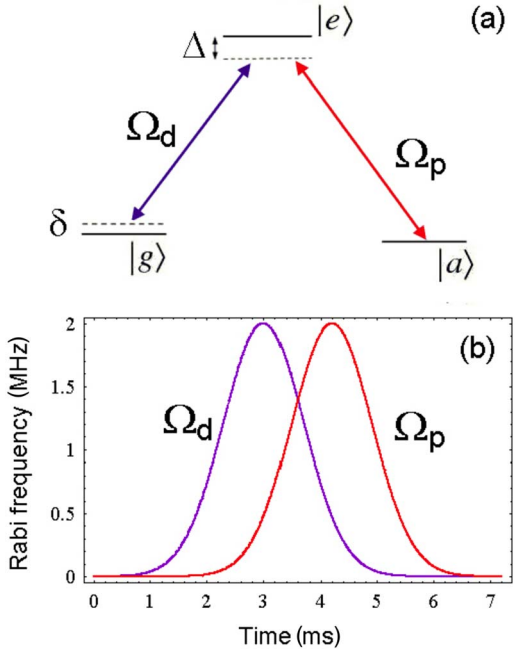

FIG. 1. (Color online). (a) A three-level Λ system. $|a\rangle$, $|e\rangle$, and $|g\rangle$ correspond to the atomic, the excited molecular, and the ground state molecular condensates. A pair of atoms from $|a\rangle$ is photoassociated in a diatomic molecule in $|e\rangle$ and is drawn into the ground state $|g\rangle$. This process in fulfilled via the counterintuitive sequence of pulses $\Omega_{p,d}$ shown in panel (b); Δ and δ indicate one-photon and two-photon detunings, respectively.

maining almost unpopulated. Therefore questions of stability and adiabaticity of the system in the dark state is essential for efficiency of population transfer. In the nonlinear (atom-molecular) STIRAP process these questions have been challenged only very recently [16,17]. Some approximate adiabatic conditions were derived, and it was also established that without mean-field collisional interactions, a system linearized about the dark state does not possess complex frequencies [17].

We are interested in nonlinear instabilities. In a recent paper [18], an improved (as compared with pioneering works of [16,17]) adiabatic condition for the nonlinear STIRAP process was derived; the instability of the system in the exact 1:1 resonance was demonstrated, and preliminary analysis of possible instabilities at 2:1 resonance between frequencies of the linearized system was performed. Both resonances turned out to be degenerate, i.e., certain conditions of generality [19] are not fulfilled.

In this paper we study the model with collisional mean-field interactions, where dynamical instabilities of the dark state due to 1:1, 2:1, and 3:1 resonances are possible. We concentrate on instabilities *not* associated with complex eigenfrequencies, since they have been rarely discussed in the BEC-related literature. Following the approach of [18], we recast the mean-field model into the form of a two-degree-of-freedom (2 DOF) classical Hamiltonian system and use the resonance normal forms theory [19,20]. In this approach the dark state corresponds to an equilibrium of the Hamiltonian system. The structure of the paper is as follows. In Sec. II, we describe the models with and without collisionless interactions, give a brief overview of the resonance normal form theory and present (for consistency) a brief discussion of 1:1 resonance in the collisionless model. In Sec. III, we discuss resonance instabilities of the dark state in the system with interactions. We show that nonlinear instabilities, besides being interesting as conceptual nonlinear phenomena, might have practical applications. In Sec. IV, we present other stationary solutions of the collisionless model. In Sec. V, concluding remarks are given, including a brief discussion of the adiabatic conditions.

II. MODELS AND METHODS

A. Models

Following [16–18], consider the three-level Λ system shown in Fig. 1(a) and briefly described in the Introduction. Neglecting collisional interactions and spontaneous emission, the mean-field equations are [16,17]

$$i\dot{\psi}_a = \Omega_p \psi_a^* \psi_e,$$

$$i\dot{\psi}_e = \Delta \psi_e + \frac{\Omega_p}{2}\psi_a^2 + \frac{\Omega_d}{2}\psi_g,$$

$$i\dot{\psi}_g = \frac{\Omega_d}{2}\psi_e, \tag{1}$$

where an overdot stands for the derivative with respect to time and the "wave functions" $\psi_{a,e,g}$ are normalized to $\sqrt{n}$ where n is the total atomic density, so that

$$|\psi_a|^2 + 2(|\psi_g|^2 + |\psi_e|^2) = 1. \tag{2}$$

We neglect spontaneous emission because mostly we are interested in motion near the dark state, but we keep in mind that it should be included in case $|e\rangle$ is populated considerably. In Eqs. (1) it is implied that two-photon detuning is zero ($\delta=0$).

Equations (1) are equivalent to Hamiltonian equations of motion of the effective classical Hamiltonian

$$H_{cl} = \frac{\Omega_p}{2}[x_2(y_1^2 - x_1^2) - 2x_1y_2y_1] - \frac{\Omega_d}{2}(x_2x_3 + y_2y_3) - \frac{\Delta}{2}(x_2^2 + y_2^2). \tag{3}$$

Here x_k are canonical momenta, while y_k are the coordinates, being related to the old "variables" (complex numbers ψ_i) as $\psi_a = x_1 + iy_1$, $\psi_e = x_2 + iy_2$, and $\psi_g = x_3 + iy_3$.

The system (1) has several stationary points, describing different physical regimes of the atomic-molecular dynamics. One of them—the dark state [16,17]—is described (up to a phase factor) by the vector $\mathbf{\Psi}_0 = (\psi_a^{(0)}, \psi_e^{(0)}, \psi_g^{(0)})^T$, where $\psi_a^{(0)} = [\frac{2\Omega_d}{\Omega_d + \Omega_e}]^{1/2}$, $\psi_e^{(0)} = 0$, $\psi_g^{(0)} = -\frac{2\Omega_p}{\Omega_d + \Omega_e}$, and $\Omega_e = \sqrt{\Omega_d^2 + 8\Omega_p^2}$. This solution is the most important in the context of the STIRAP process (as it has zero population of the excited state). Some other stationary states that might be important for other applications are given in Sec. IV.

To include two-body interactions in the STIRAP process, one introduces a two-photon detuning δ in the model in order to "compensate" for nonlinearities and keep the dark state as a stationary solution of the system [21]. That is, the equations of motion are

$$i\dot{\psi}_a = f_a \psi_a + \Omega_p \psi_a^* \psi_e,$$

$$i\dot{\psi}_e = (f_e + \Delta)\psi_e + \frac{\Omega_p}{2}\psi_a^2 + \frac{\Omega_d}{2}\psi_g,$$

$$i\dot{\psi}_g = (f_g + \delta)\psi_g + \frac{\Omega_d}{2}\psi_e, \tag{4}$$

where $f_{a,e,g}$ are nonlinear terms coming from collisional interactions ($f_i = \Sigma g_{ij}|\psi_j|^2$, g_{ij} are proportional to the s-wave scattering lengths of atom-atom, atom-molecule, and molecule-molecule collisions [17]). Exactly the same method that was used in [18] for the system (1) without interactions can be applied to the system (4) with interactions. This is an important finding of the present paper, since presently available methods [17] lead to heavy complications when interactions are taken into account. We take into account atom-atom collisional interactions and interactions involving molecules in the ground state, i.e., we neglect only collisions involving molecules in the excited states. Corresponding coefficients are g_{aa} (for atom-atom), g_{gg} (ground state molecule-molecule), and g_{ag} (atom-ground state molecule). From Eqs. (4) we obtain an equivalent classical Hamiltonian

$$H_{int} = H_{cl} - \frac{\delta}{2}(x_3^2 + y_3^2) - \frac{g_{aa}}{4}(x_1^2 + y_1^2)^2 - \frac{g_{gg}}{4}(x_3^2 + y_3^2)^2 - \frac{g_{ag}}{2}(x_1^2 + y_1^2)(x_3^2 + y_3^2), \quad (5)$$

where, according to [21], detuning δ should be chosen to "compensate" for the nonlinearities:

$$\delta = (2g_{aa} - g_{ag})|\psi_a^{(0)}|^2 + (2g_{ag} - g_{gg})|\psi_g^{(0)}|^2 = \frac{4\Omega_d(2g_{aa} - g_{ag}) + (2g_{ag} - g_{gg})(\Omega_e - \Omega_d)}{2(\Omega_e + \Omega_d)}. \quad (6)$$

As a result, the absolute values of the components of the dark state remain unchanged, but the phases of ψ_a and ψ_g now rotate with the frequencies $\mu = g_{aa}|\psi_a^{(0)}|^2 + g_{ag}|\psi_g^{(0)}|^2$ and 2μ, correspondingly: $\psi_a^{(0)} = |\psi_a^{(0)}|e^{-i\mu t}$, $\psi_e^{(0)} = 0$, $\psi_g^{(0)} = |\psi_g^{(0)}|e^{-2i\mu t}$.

B. Stability of Hamiltonian systems and resonance normal form theory

The eigenfrequencies of the system linearized about the dark state are given by [17]: $\omega_0 = 0$ and $\omega_\pm = \frac{1}{2}[\Delta \pm (\Delta^2 + \Omega_d\Omega_e)^{1/2}]$. They are real which, however, does not guarantee yet that the state is dynamically stable: there may occur nonlinear instabilities. The study of such instabilities is performed using the theory of normal forms (see, e.g., [19]), which for the sake of convenience we outline below for the case of a system with two degrees of freedom, i.e., for the case we are dealing with in this paper.

The first step consists in reduction of the original system to the 2 DOF Hamiltonian system, whose equilibrium point located at the origin corresponds to the dark state. Next, in the vicinity of the equilibrium, one considers in the first approximation only the quadratic terms of the Hamiltonian function:

$$H \approx H_2 = \frac{1}{2}(Az, z), \quad (7)$$

where z designates a vector of canonical coordinates and momenta and A is a 2×2 square matrix, so that the equations of motion have the form

$$\dot{z} = I\frac{\partial H_2}{\partial z} = Az, \quad (8)$$

where $I = \begin{pmatrix} 0 & -E_2 \\ E_2 & 0 \end{pmatrix}$, and hereafter E_m stands for the $m \times m$ unity matrix. Eigenvalues of the Hamiltonian H_2 are the roots of the equation $\det(IA - \lambda E_4) = 0$. In the complex plain the roots are located symmetrically about the coordinate cross, i.e., if λ is an eigenvalue then $\bar{\lambda}, -\lambda, -\bar{\lambda}$ are the eigenvalues of H_2, too. As a result, the stability of a Hamiltonian system is always neutral: if an equilibrium is stable, then real parts of all the eigenvalues are equal to zero.

Instabilities in Hamiltonian systems is not a trivial issue, as they do not necessarily arise from complex eigenfrequencies, which correspond to complex eigenvalues. Eigenvalues of H_2 can be of four different types [19]: real pairs $(a, -a)$, purely imaginary pairs $(ib, -ib)$, complex double pairs $(\pm a \pm ib)$, and zero eigenvalues. By means of a linear canonical transformation, the quadratic Hamiltonian can be reduced to a sum of partial Hamiltonians (i.e., functions of nonintersecting sets of canonically conjugated variables), where every partial Hamiltonian corresponds to one of the above-mentioned four types of eigenvalues.

Suppose that all eigenvalues are purely imaginary and different, then the Hamiltonian H_2 can be reduced to a normal form $H_2 = \omega_1(p_1^2 + q_1^2)/2 + \omega_2(p_2^2 + q_2^2)/2$ and the full Hamiltonian H in the vicinity of the equilibrium can be represented as $H = H_2 + H_3 + H_4 + \cdots$, where H_j $(j = 3, 4, \ldots)$ is a homogeneous polynomial of the jth order.

Now, recalling that we are dealing with two degrees of freedom, the equilibrium will be stable if there are no resonances up to the fourth order between the frequencies $\omega_{1,2}$ (the order of a resonance relation $k_1\omega_1 + k_2\omega_2 = 0$ is defined as $k \equiv |k_1| + |k_2|$), and the so-called isoenergetic nondegeneracy condition [19] is fulfilled. Thus in a vicinity of a low-order resonance, careful examination of the Hamiltonian is needed, including analysis of higher-order terms (i.e., H_3 and H_4). In the next paragraph we briefly analyze 1:1 resonance in the collisionless model.

C. 1:1 resonance in the collisionless model

The collisionless model is very useful for demonstrating nonlinear instability: there is only one type of nonlinearity, and linear instabilities are absent. The system determined by classical Hamiltonian (3) has two degrees of freedom only because of the integral (2) which can be rewritten as $x_1^2 + y_1^2 + 2(x_2^2 + y_2^2 + x_3^2 + y_3^2) = 1$. There is a set of stationary points filling a circumference $x_1^2 + y_1^2 = 2\Omega_d/(\Omega_e + \Omega_d) = A^2$. This circumference corresponds to the dark state: its radius corresponds to the normalized population of $|a\rangle$, and position on the circumference to the phase of ψ_a. Introducing polar coordinates $x_k = \sqrt{2\rho_k}\cos\phi_k$ and $y_k = \sqrt{2\rho_k}\sin\phi_k$ $(k = 1, 2, 3)$ and subsequently defining $p_m = \sqrt{2\rho_{m+1}}\cos(\phi_{m+1} - 2\phi_1)$ and $q_m = \sqrt{2\rho_{m+1}}\sin(\phi_{m+1} - 2\phi_1)$ $(m = 1, 2)$ [i.e., $(p_{2,3}, q_{2,3})$ are the Cartesian coordinates $(x_{2,3}, y_{2,3})$ rotated by the angle $2\phi_1 = 2\arctan(x_1/y_1)$], we reduce H_{cl} [Eq. (3)] to the following 2 DOF Hamiltonian [22]:

$$H_{pq} = \Omega_p\left[(q_1^2 + p_1^2 + q_2^2 + p_2^2) - \frac{1}{2}\right]p_1 - \frac{\Omega_d}{2}(q_1q_2 + p_1p_2) - \frac{\Delta}{2}(q_1^2 + p_1^2). \quad (9)$$

The stationary point of H_{pq} that corresponds to the dark state circumference is $p_1 = q_1 = q_2 = 0, p_2 = \frac{\Omega_d - \Omega_e}{4\Omega_p} = B$. Shifting the origin to that point, we get the Hamiltonian

$$\mathcal{H} = \Omega_p(q_1^2 + p_1^2 + q_2^2 + p_2^2)p_1 - \frac{\Omega_d}{2}\left(q_1q_2 + \frac{\Omega_e}{\Omega_d}p_1p_2\right) - \frac{\Delta}{2}(q_1^2 + p_1^2), \quad (10)$$

where now p_2 is the deviation from the stationary point.

Eigenfrequencies of the system linearized about the origin are equal to $\omega_{\pm}$, therefore at $\Delta=0$ 1:1 resonance happens: $\omega_+ + \omega_- = 0$ [18]. After a series of transformations described in [18], one obtains an effective Hamiltonian

$$F = \frac{1}{2}J - \frac{J\kappa^2}{8}\left[\frac{1}{2}\left(P^2 + \frac{J^2}{R^2}\right)A_P + R^2 A_R\right], \qquad (11)$$

where $A_P=(10\cos^2\theta+10\cos\theta+4)/3$ and $A_R=5\cos\theta-1$, the parameter θ is defined as parameter θ as $\Omega_d=\Omega_e\cos\theta$.

The idea behind the transformations is to represent the Hamiltonian in the vicinity of the origin as $H=H_2+H_3+H_4$, get rid of the cubic terms H_3 by means of a nonlinear canonical transformation, and then average the resulting Hamiltonian over a variable which is "fast" due to 1:1 resonance (this variable is canonically conjugated to J in Eq. (11), as a result $J=$const after the averaging) [18].

The Hamiltonian (11) is an integrable system for a pair of canonically conjugated variables P,R. It is not difficult to understand its dynamics: Eq. (11) is merely a Hamiltonian of a particle in the effective potential $U(R)=\frac{J^2A_P}{2R^2}+A_R R^2$ [indeed, only the expression in the square brackets in Eq. (11) is important for dynamics]. For $\cos\theta>\frac{1}{5}$ (i.e., for $\Omega_p<\sqrt{3}\Omega_d$), the system (11) has a single fixed point, while for $\cos\theta<\frac{1}{5}$ there are no fixed points [see Fig. 2(a)]. In the latter case, a phase point initially placed close to $R=0$ will slowly move from the origin to large values of R. The dark state corresponds to $R=0$, so that such an effective potential implies instability of the dark state due to 1:1 resonance. Numerical examples are presented in Fig. 2(b). Physically, $\cos\theta<1/5$ means that most of the population is in the ψ_g state. Thus we have an interesting counterintuitive phenomenon: *the dark state becomes unstable provided* $\Omega_p>\sqrt{3}\Omega_d$: deviations from it slowly grow with time.

Let us now briefly recall the generic behavior of a Hamiltonian system near the 1:1 resonance. The 1:1 resonance means that eigenvalues of a linearized Hamiltonian collide. Usually, in the vicinity of the resonance one brings a quadratic part of the Hamiltonian to the normal form $H=a(p_1^2+p_2^2)+b(p_2q_1-p_1q_2)+\tilde{\delta}(q_1^2+q_2^2)$, where $\tilde{\delta}$ is a deviation from the resonance. In the nondegenerate case ($a\neq 0$), an exponential instability can arise due to the quadratic part: at one side of the resonance ($\tilde{\delta}<0$), instability is exponential in time, because the eigenvalues $\lambda_i=\pm(ib\pm 2i\sqrt{a\tilde{\delta}})$ leave the imaginary axis ("split") after the collision at $\tilde{\delta}=0$. At the other side of the resonance, the eigenvalues are imaginary (i.e., the eigenfrequencies are purely real); nevertheless in the vicinity of the resonance equilibrium may become unstable due to higher-order terms [19]. In our case, even when the detuning from the resonance $\tilde{\delta}=\Delta/2$ is introduced, the eigenvalues cannot leave the imaginary axis: they "pass" each other when $\tilde{\delta}$ is changed; instead of a nonsemisimple matrix A [see Eq. (8)], we have a diagonizable matrix. As we clarified above, the instability comes from the quartic terms of the Hamiltonian being transformed to the normal form.

Since long time scales are required, for real applications it may be necessary to take into account spontaneous emission. In our classical approach, it will enter as a dissipation. We postpone this investigation to future work. Here we add some notes on 2:1 resonance. Analysis of [18] showed that this resonance is also degenerate (the resonance term is merely absent). Similar to 1:1 resonance, we need to analyze the quartic terms here, which is not an easy task (see, for example, [30]).

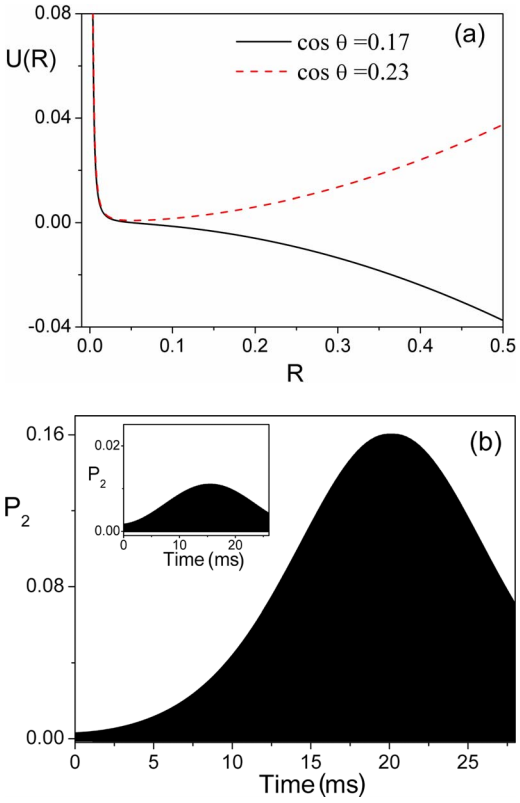

FIG. 2. (Color online). (a) The effective potential $U(R)$ of the Hamiltonian (11) for $\cos\theta=0.17$ (the solid line) without a local minimum and for $\cos\theta=0.23$ (the dashed line) where a stable equilibrium close to the origin (i.e., to the dark state) exists. (b) The evolution of the population P_2 of the excited state, starting with a small deviation from the dark state (for which P_2 is exactly zero). The parameters are $\Omega_p=2.0\times10^6$ s^{-1} and $\Omega_d=1.0\times10^6$ s^{-1} ($\cos\theta\approx0.17$). We see that instability develops on long time scales (10 ms); deviations from the dark state grow and reach very large values [notice that the maximum value of P_2 is 0.5 due to the normalization $P_1+2(P_2+P_3)=1$]. In the inset the dynamics with $\Omega_p=1.5\times10^6$ s^{-1}, $\Omega_d=1.0\times10^6$ s^{-1} ($\cos\theta\approx0.23$) is shown. Here, the effective potential has a minimum, so deviations from the dark state first grow with time, and then begin to oscillate slowly about this effective equilibrium. Note that the amplitude of these oscillations is an order of magnitude smaller than in the unstable case.

III. RESONANCES IN THE MODEL WITH TWO-BODY INTERACTIONS

A. Mathematical analysis

In [21] it was suggested that the STIRAP process should be modified in the presence of collisional interactions. In that case the two-photon detuning δ (see Fig. 1) should be introduced to compensate the effect of the nonlinearity, leading to

a more complicated system (4) with the constraint (6) on the value of δ. As we have already mentioned above, the model (4) is equivalent to the classical Hamiltonian H_{int} in Eq. (5). This Hamiltonian is rather complicated, as it contains even quartic terms. We may fulfill the analysis of resonant instabilities using the same approach as in the previous section. By making the respective canonical transformations we get instead of Eq. (9) the following 2 DOF Hamiltonian:

$$H_{pq}^{int} = \Omega_p\left(q_1^2 + p_1^2 + q_2^2 + p_2^2 - \frac{1}{2}\right)p_1 - \frac{\Omega_d}{2}(q_1q_2 + p_1p_2) - \frac{\Delta}{2}(q_1^2 + p_1^2) - \frac{\delta}{2}(p_2^2 + q_2^2) - g_{aa}\left(q_1^2 + p_1^2 + q_2^2 + p_2^2 - \frac{1}{2}\right)^2 - \frac{g_{gg}}{4}(p_2^2 + q_2^2)^2 - g_{ag}(p_2^2 + q_2^2)\left(\frac{1}{2} - (q_1^2 + p_1^2 + q_2^2 + p_2^2)\right). \quad (12)$$

Next, shifting the origin to the fixed point corresponding to the dark state, we obtain $H=H_2+H_3+H_4$ where [cf. Eq. (10)]

$$H_2 = -\frac{\Omega_d}{2}q_1q_2 - \frac{\Omega_e}{2}p_1p_2 - \frac{\bar{\Delta}}{2}(q_1^2 + p_1^2) - g_Bp_2^2, \quad (13)$$

H_3 and H_4 designate cubic and quartic polynomials in the new variables $q_{1,2}, p_{1,2}$,

$$\bar{\Delta} \equiv \Delta - 2g_{aa}|\psi_a^0|^2 - 2g_{ag}|\psi_g^0|^2 = \Delta - 2\mu,$$

$$g_B = (4g_{aa} - 4g_{ag} + g_{gg})|\psi_g^0|^2 = \frac{16D_g\Omega_p^2}{(\Omega_e + \Omega_d)^2},$$

and $D_g = g_{aa} - g_{ag} + g_{gg}/4$.

Let us consider H_2 given by Eq. (13) in more detail. Assume its eigenvalues are purely imaginary. Then one can transform H_2 to a sum of two linear oscillators. Skipping the corresponding sequence of canonical transformations, the resulting Hamiltonian has the form

$$H_2 = \frac{1}{2}\tilde{\omega}_1(P_1^2 + Q_1^2) + \frac{1}{2}\tilde{\omega}_2(P_2^2 + Q_2^2), \quad (14)$$

where

$$\tilde{\omega}_1 = -\frac{1}{2}\sqrt{2\bar{\Delta}^2 + \Omega_{de}^2 + 2\sqrt{\bar{\Delta}^4 + \bar{\Delta}(\bar{\Delta} + 2G)\Omega_{de}^2}},$$

$$\tilde{\omega}_2 = \frac{1}{2}\sqrt{2\bar{\Delta}^2 + \Omega_{de}^2 - 2\sqrt{\bar{\Delta}^4 + \bar{\Delta}(\bar{\Delta} + 2G)\Omega_{de}^2}}, \quad (15)$$

$G = g_B\Omega_d/\Omega_e$, and $\Omega_{de} = \sqrt{\Omega_d\Omega_e}$. Let us list possible low-order resonances ($k_1:k_2$ resonance below means $k_1\tilde{\omega}_1 + k_2\tilde{\omega}_2 = 0$).

$$1{:}1 \quad \bar{\Delta} = 0,$$

$$1{:}2 \quad 64\bar{\Delta}^2(\bar{\Delta}^2 + \Omega_{de}^2) - \Omega_{de}^2(9\Omega_{de}^2 - 200\bar{\Delta}G) = 0, \quad (16)$$

$$1{:}3 \quad 36\bar{\Delta}^2(\bar{\Delta}^2 + \Omega_{de}^2) - \Omega_{de}^2(16\Omega_{de}^2 - 200\bar{\Delta}G) = 0,$$

$$1{:}0 \quad \Omega_{de}^2 = 8G\bar{\Delta}.$$

These resonances can lead to loss of stability of the dark state even if cases $\tilde{\omega}_{1,2}$ are purely real. Let us consider in detail the 1:1 resonance in order to have the full picture of this highly nontrivial degeneracy in both the collisionless system and the system with interactions.

Assume the exact resonance $\bar{\Delta}=0$. The quadratic part of the Hamiltonian acquires the form

$$\mathcal{H}_2 = -\frac{\Omega_{de}}{2}(q_1q_2 + p_1p_2) - Gp_2^2. \quad (17)$$

The matrix A of the corresponding equations of motion [see Eq. (8)] is nonsemisimple (i.e., not diagonalizable: its Jordan decomposition has two Jordan blocks of the second order). One, however, can use the general approach to the 1:1 resonance [19]. Following this way, we make a sequence of (rather involved and requiring symbolic computer calculations) canonical transformations bringing the full Hamiltonian to the standard normal form (see also the Appendix):

$$H = \frac{1}{2}a\left(P^2 + \frac{J^2}{R^2}\right) + J\omega + R^2\left(DR^2 + FJ + C\left(P^2 + \frac{J^2}{R^2}\right)\right), \quad (18)$$

where the coefficients F and C depend on $\Omega_{p,d}, g_{aa}, g_{ag}, g_{gg}$ in a complicated way, while the most essential parameters a and D can be represented by rather simple expressions, with the help of the angle parameter θ defined as $\cos\theta = \Omega_d/\Omega_e$:

$$a = -2D_g\cos\theta\tan^2\frac{\theta}{2},$$

$$D = \frac{1}{64}[8d_g - 35D_g - 6g_{aa} + (48D_g - 10d_g)\cos\theta - 19D_g\cos 2\theta].$$

Here $d_g = 2g_{aa} - g_{ag}$.

The analysis of the phase portraits of Eq. (18) is available, e.g., in [19]. In Fig. 3 we present some typical examples for $aD<0$ [panels (a) and (b)] and for $aD>0$ [panel (c)]. At $aD<0$, there are regions of instability (runaway trajectories) on the phase portraits.

The origin (i.e., dark state equilibrium) is stable in the case $aD>0$ and is unstable in the case $aD<0$ [19]. The term with C in Eq. (18) is not so important for dynamics, as it is much smaller than the first term in Eq. (18) (note that the two terms differ by the small factor R^2). The term with F may create a region of stability near equilibrium (at finite J); however, at some larger distances from the equilibrium dynamics is determined by coefficients a and D (see detailed bifurcation diagram in [19]). The key magnitude for stability of the dark state is therefore

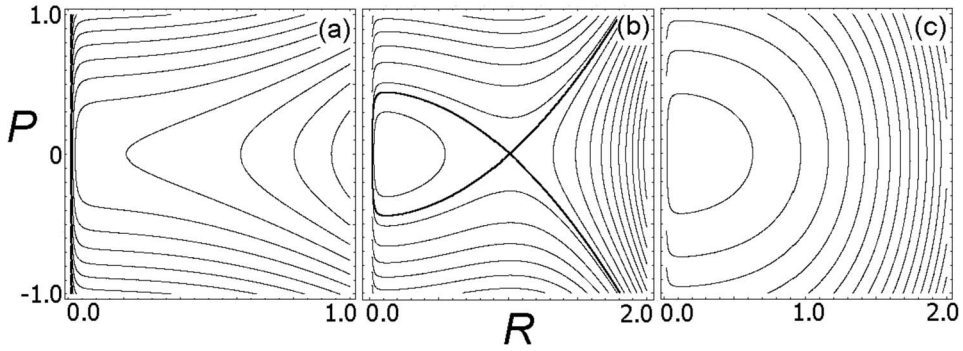

FIG. 3. Typical phase portraits of the normal form (18) in the vicinity of 1:1 resonance. In (a) and (b) $aD<0$, while in (c) $aD>0$. The dark state equilibrium corresponds to the origin of the phase portraits ($P=0$, $R=0$), so the runaway trajectories at $aD<0$ imply instability of the equilibrium.

$$aD = -\frac{D_g}{32}[8d_g - 35D_g - 6g_{aa} + (48D_g - 10d_g)\cos\theta - 19D_g\cos 2\theta]\cos\theta\tan^2\frac{\theta}{2}. \quad (19)$$

This is one of the main analytical results of the paper. In Sec. III B we discuss its physical implications.

B. Physical discussion

Let us now turn to the discussion of the physical consequences of the obtained results. We start with the observation of a very important feature of the expression (19): while $aD \sim n^2$ (n is the density of the condensate), its qualitative behavior does not depend neither on the density nor on the absolute values of $\Omega_{p,d}$: it only depends on the scattering lengths and on the parameter θ, as it will be described below. This suggests a possible application of the resonances: they can be employed for precise determination of the scattering lengths.

For the estimates of the relevant times we use the same set of experimentally feasible parameters as in Ref. [21], i.e., we consider a ^{87}Rb condensate with the density $n=4.3\times10^{20}$ m^{-3}, what corresponds to [21]: $g_{aa}=21\,328$ s^{-1}, $g_{ag}=-27\,692$ s^{-1}, and $g_{gg}=10\,664$ s^{-1}. In Fig. 4 we illustrate typical dependences of aD vs $\cos\theta$ for these values of the parameters. From the figure one observes that the parameter aD becomes negative in a narrow range of the parameter θ given by $0.4\lesssim\cos\theta\lesssim0.5$, which implies the existence of two different types of behavior inside and outside of this window. This analytical prediction is supported by the direct numerical simulations shown in Fig. 5. At either $\cos\theta<0.4$ or $\cos\theta>0.5$ small deviations from the dark state do not grow appreciably [see panels (a) and (d)]. If, however, $\cos\theta\in[0.4,0.5]$, initial small deviation may grow up to very high values. We also notice that initially small population P_2 grows even if θ is taken slightly outside the window $\cos\theta\in[0.4,0.5]$. This means that in the effective potential a stable fixed point is located at a certain (small) distance from the origin (see Fig. 3). However, as is expected the dramatic growth of amplitude of the excited state population is observed when the parameter θ is inside the instability window $\cos\theta\in[0.4,0.5]$. Say, for $\cos\theta=0.4$ [Fig. 5(c)] the maximal amplitude of P_2 is an order of magnitude higher than for $\cos\theta=0.2$ [Fig. 5(d)]. For $\cos\theta=0.5$, we also give a numerical example [see inset of Fig. 5(b)] where initial conditions are exactly as in Fig. 5(b), but a value of Δ was chosen to be 10% higher (i.e., $\bar{\Delta}\neq0$, and we are far from 1:1 resonance). With such a detuning from the resonance, we see the dramatic decrease of the amplitude of oscillations of the population P_2: the observed increase is three orders of magnitude. It can be seen also that the time scale for the development of the nonlinear instability is of order of milliseconds, i.e., is feasible experimentally. It is important to emphasize that the time scales depend not only on the parameters of the system, but also on the initial population of the excited state. When initially the system is prepared far enough from the equilibrium (i.e., far from the dark state), the instability develops much faster—on time scales of hundreds or even tens of microseconds.

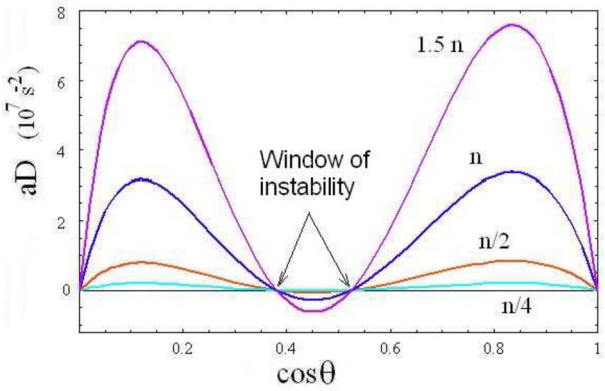

FIG. 4. (Color online). Dependence of the parameter aD on $\cos\theta$ as given by Eq. (19). We consider the same example as in [21]: ^{87}Rb atoms with density $n=4.3\times10^{20}$ m^{-3}, which gives the following values of parameters [21]: $g_{aa}=21\,328$ s^{-1}, $g_{ag}=-27\,692$ s^{-1}, and $g_{gg}=10\,664$ s^{-1}. A region of the instability of the dark state due to 1:1 resonance corresponds to $aD<0$. Several over values of density are depicted as well ($1.5n, n/2, n/4$). While magnitude of aD depends on density ($aD\sim n^2$), its qualitative behavior only depends on θ and values of the scattering lengths. The resonant behavior therefore might be used for precise determination of the scattering lengths.

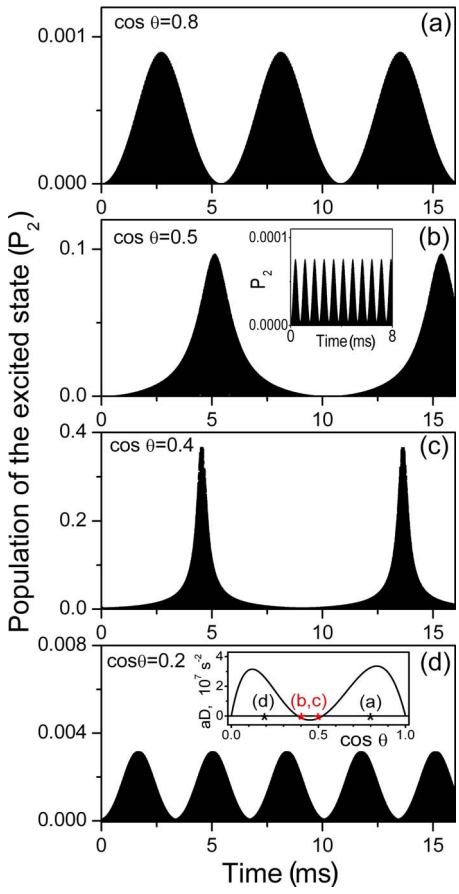

FIG. 5. (Color online). Time evolution of the population P_2 starting with the initial value $P_2(t=0)=10^{-6}/\cos\theta$ for different θ. (Notice that while we show only P_2, deviations of the other components from its dark state values are of the same order, i.e., initially of the order of 10^{-6}.) We illustrate three different types of behavior: small oscillations in the vicinity of 1:1 resonance, when parameter θ lies outside the instability window shown in Fig. 4 [panels (a) and (d)], huge oscillations in the instability window [panels (b) and (c)], and tiny oscillations far from the 1:1 resonance [inset in panel (b)]. In all the figures [except the inset of panel (b)] the parameters were chosen to satisfy the resonance condition $\bar{\Delta}=0$ exactly: we fixed $\Omega_p=2.1\times10^6$ s^{-1} and determined Ω_d by the value of $\cos\theta$. For the inset in panel (b) all initial conditions are the same as in (b), but the detuning Δ was chosen to be 10% higher than its value in (b). As a result, the system is far from 1:1 resonance ($\bar{\Delta}\neq0$), and the amplitude of oscillations is dramatically decreased (in three orders of magnitude). The inset of (d) shows $aD(\cos\theta)$ from Fig. 4; values of $\cos\theta$ in plots (a)–(d) are marked by asterisks.

IV. OTHER SOLUTIONS OF THE ATOM-MOLECULE SYSTEM

Apart from the dark state, the system (1) has several other stationary solutions, which might be important in future experimental techniques. In order to present them we introduce a positive parameter Ω_0 defined through the relation $\Omega_0^2=\Delta^2+\Omega_d^2$. Then the state

$$\Psi_1=\begin{pmatrix}\psi_a^{(1)}\\ \psi_e^{(1)}\\ \psi_g^{(1)}\end{pmatrix}=\frac{1}{4\Omega_p}\begin{pmatrix}2\sqrt{\mu_0^2-\Omega_0^2}e^{-i(\mu_0+\Delta)t/4}\\ (\mu_0+\Delta)e^{-i(\mu_0+\Delta)t/2}\\ \Omega_d e^{-i(\mu_0+\Delta)t/2}\end{pmatrix}, \tag{20}$$

where $\mu_0^2>\Omega_0^2$, μ_0 being a real parameter, is a solution of Eqs. (1). The specific value of μ_0 is found from the normalization condition (2):

$$\mu_0=-\frac{\Delta}{3}-\frac{1}{3}\sqrt{4\Delta^2+3\Omega_d^2+24\Omega_p^2} \tag{21}$$

and is supplied by the constraint $\mu_\pm^2>\Omega_0$, which is equivalent to

$$\sqrt{4\Delta^2+24\Omega_p^2+3\Omega_d^2}-2\Delta\leqslant3\sqrt{8\Omega_p^2-\Omega_d^2}. \tag{22}$$

As it is evident, the last formula can always be satisfied by choosing large enough single-photon detuning Δ and by requiring $8\Omega_p^2>\Omega_d^2$ which can be achieved, say, by increasing the atomic density. We leave the stability of this state as well as the effect of the spontaneous emission on it for further study.

V. CONCLUDING REMARKS

To summarize the findings of the present paper we have shown that the method of description of the nonlinear instabilities suggested in [18] can be successfully used for the atomic molecular condensates with intra- and inter-species two-body interactions. We found nonlinear instabilities due to 1:1, 1:0, 2:1, and 3:1 resonances in the Λ system with collisional interactions. The instabilities may be important for manipulations of the conversion processes that occur on relatively long time scales.

Following the earlier work [18] on 1:1 resonance in the collisionless model, the present paper provides a detailed discussion of the nontrivial degeneracy in the model with and without collisional interactions. In the collisionless model, the two eigenvalues of the linearized system always remain on the imaginary axis. By changing the detuning Δ one moves the eigenvalues along the axis. In the exact resonance $\Delta=0$, the equations of motion are determined by a semisimple (diagonizable) matrix A. Analysis of the nonlinear terms [18] shows that there exists instability in a certain range of the parameters ($\cos\theta<1/5$, i.e., $\Omega_p>\sqrt{3}\Omega_d$).

In the model with two-body collisional interactions the situation is different. The eigenvalues can leave the imaginary axis. On one side of the resonance (with the effective detuning $\bar{\Delta}<0$) the eigenvalues become complex, which lead to the "usual" exponential instability. In the exact resonance the matrix A is nonsemisimple, and the general approach to 1:1 resonance [19] is to be used. The equilibrium turns out to be either stable or unstable depending on the value of the parameter aD. Note that at a certain stage (in the analysis of quartic terms) we use averaging instead of a standard normal form approach [19,20,27,28,30]; the two approaches are mathematically equivalent, but the former is simpler from the point of view of symbolic calculations.

The analysis developed above implies time-independent parameters (i.e., Rabi frequencies $\Omega_{p,d}$ and the detunings δ,Δ). With time-dependent parameters (corresponding to the STIRAP process), at least two important issues arise. The first one is the adiabatic conditions. In case the system is sufficiently far away from the low-order resonances mentioned above, one can generalize the adiabatic condition of Ref. [18] straightforwardly. That is, in the vicinity of an instantaneous dark state the system can be approximated by two linear oscillators with time-dependent frequencies ω_k, $k=1,2$. Change of the frequency during one period of the unperturbed motion should be much less than the frequency itself, which provides us with simple adiabatic conditions $\dot{\omega}_k \ll \omega_k^2/2\pi$. In the vicinity of low-order resonances, however, the linearized system is not enough even to determine stability of the system at given parameters (it is determined by the nonlinear terms). Therefore, even at perfect adiabatic conditions, that is at constant parameters, the system may go far away from the dark state.

The second relevant issue is the passage through a resonance [31,32]. At a static situation, the resonances are not so impressive. The most interesting phenomena happen when the parameters are changed in such a way that the system passes through the region in its phase space where the resonance conditions are approximately fulfilled (the resonance region). At passage through the resonance region, scattering on resonance and capture into a resonance may happen [31,32]. Passage through the resonance is in some sense a less complicated issue than *passage through the separatrix* [19,23–25] (i.e., "separatrix crossing," which was already studied in relation to BEC physics [26]). These topics will be discussed in detail elsewhere. We emphasize that the present study, in particular, opens a way to implement in an atomic-molecular BEC an interesting and unusual technique of control of nonlinear systems: the capture into a resonance [19,31,32].

Subsequent manuscripts [29] study in detail improved adiabatic conditions of the nonlinear STIRAP process and passages through the resonances. The approach suggested here and in [18] can be used for many other nonlinear BEC systems. For example, BEC tunneling in a three-dimensional lattice [33] could be mentioned.

ACKNOWLEDGMENTS

A.P.I. was supported by JSPS and 21st Century COE program on "Coherent Optical Science." This work was also supported by Grants-in-Aid for Scientific Research No. 16-04315 from the Ministry of Education, Culture, Sports, Science and Technology, Japan. V.V.K. was supported by Grant No. POCI/FIS/56237/2004 (FCT Portugal and European program FEDER). A.P.I. gratefully acknowledges an encouraging discussion with E. Arimondo, and many clarifying discussions with A.A. Vasiliev and A.I. Neishtadt. It is a great pleasure for A.P.I. to thank J. Bohn for his kind invitation to visit JILA at Boulder, CO, where part of this work was done. The authors thank S. Dmitriev for discussions and encouragement.

APPENDIX: TRANSFORMATION OF THE HAMILTONIAN (13) TO THE NORMAL FORM

Consider exact 1:1 resonance $\bar{\Delta}=0$ for simplicity. For the sake of convenience, we start with the transformation $(q_1,p_1)\to(\tilde{p}_1,-\tilde{q}_1)$, $(q_2,p_2)\to(\tilde{q}_2\sqrt{\Omega_e/\Omega_d})$, $\tilde{p}_2\sqrt{\Omega_d/\Omega_e})$. Then, the explicit expression for the parts of the Hamiltonian (13) have the following form (omitting tildes over the new variables):

$$H_2=-\frac{\Omega_{de}}{2}(p_1q_2-p_2q_1+Gp_2^2),$$

$$H_3=\frac{4d_g\Omega_p\sqrt{\Omega_d}p_1^2p_2}{\sqrt{\Omega_e}(\Omega_d+\Omega_e)}+\frac{8D_g\Omega_p\Omega_d^{3/2}p_2^3}{\Omega_e^{3/2}(\Omega_d+\Omega_e)}-\Omega_pp_1^2q_1-\frac{\Omega_d\Omega_pp_2^2q_1}{\Omega_e}+\frac{4d_g\Omega_p\sqrt{\Omega_d}p_2q_1^2}{\sqrt{\Omega_e}(\Omega_d+\Omega_e)}-\Omega_pq_1^3+\frac{8D_g\Omega_p\sqrt{\Omega_e}p_2q_2^2}{\sqrt{\Omega_d}(\Omega_d+\Omega_e)}-\frac{\Omega_e\Omega_pq_1q_2^2}{\sqrt{\Omega_d}},$$

$$H_4=-g_{aa}p_1^4-2g_{aa}p_1^2q_1^2-\frac{D_g\Omega_d^2p_2^4}{\Omega_e^2}-2g_{aa}p_1^2q_1^2-\frac{d_g\Omega_dp_2^2q_1^2}{\Omega_e}-g_{aa}q_1^4-\frac{d_g\Omega_ep_1^2q_2^2}{\Omega_d}-2D_gp_2^2q_2^2-\frac{d_g\Omega_eq_1^2q_2^2}{\Omega_d}-\frac{D_g\Omega_e^2q_2^4}{\Omega_d^2},$$

where $D_g=g_{aa}-g_{ag}+g_{gg}/4$ and $d_g=2g_{aa}-g_{ag}$.

We eliminate the cubic terms (i.e., H_3) by means of a canonical transformation

$$W_3=a_1P_2^2q_2+a_2q_1^2q_2+a_3P_2q_2q_1+a_4P_1P_2^2+a_5P_1P_2q_1+a_6P_1^3+a_7q_1^2P_1+a_8q_2^3+a_9P_1q_2^2+a_{10}P_1^2q_2, \quad \text{(A1)}$$

where explicit expressions for the coefficients $(a_1,\ldots,a_{10})$ are obtained using MATHEMATICA and are too cumbersome to be presented explicitly. Up to terms of the second order, the new and old variables are related by

$$p_k=P_k+\frac{\partial\bar{W}_3}{\partial Q_k},\quad q_k=Q_k-\frac{\partial\bar{W}_3}{\partial P_k},\quad k=1,2, \quad \text{(A2)}$$

where $\bar{W}_3\equiv W_3(P_k,Q_k)$, i.e., the function $\bar{W}_3$ is obtained from W_3 by replacing old coordinates q_k with the new ones (Q_k). The transformation generated by W_3 kills all cubic terms, while H_2 remains the same. To investigate the quartic terms, we need to determine the relations between the new and old variables with the accuracy up to the third order in P_k,Q_k. It can be done recursively [replacing Q_k in the right-hand side of Eq. (A2) by more accurate expressions, given by Eq. (A2) itself].

Then, we bring the quadratic part H_2 to the normal form by means of a linear canonical transformation $p_{1,2}=P_1\mp P_2$, $q_{1,2}=(\pm GP_2+Q_1\mp Q_2)/2$. The quadratic part takes the form $\mathcal{H}_2=\frac{a(P_1^2+P_2^2)}{2}+\frac{\Omega_{de}}{2}(P_2Q_1-P_1Q_2)$, while the quartic part can be further reduced to the form

$$\mathcal{H}_4 = (Q_1^2 + Q_2^2)[D(Q_1^2 + Q_2^2) + B(P_2Q_1 - P_1Q_2) + C(P_1^2 + P_2^2)]. \quad \text{(A3)}$$

We change to polar coordinates in the Q_1, Q_2 plane by means of a transformation

$$Q_1 = R\cos\xi, \quad Q_2 = R\sin\xi, \quad P_1 = P\cos\xi - \frac{J}{R}\sin\xi,$$

$$P_2 = P\sin\xi + \frac{J}{R}\cos\xi, \quad \text{(A4)}$$

and then average over the angle ξ, which finally leads us to the explicit form (18).

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