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Demkov–Kunike Model in Cold Molecule Formation: the Fast Resonance Sweep Regime of the Strong Interaction Limit

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Abstract—We study a quadratic-nonlinear version of the level-crossing Demkov–Kunike problem relevant to coherent molecule formation via photo- or Feshbach-association of ultracold atoms. Using an exact third-order nonlinear differential equation for the molecular state probability, we construct an approximate solution to the problem in the fast sweep regime of the strong interaction limit. The constructed approximation, defined as a solution of a first-order nonlinear equation, contains a fitting parameter which we determine through a variational procedure. The suggested approach can be interpreted as a modification of the adiabatic approximation and can further be used for the treatment of analogous nonlinear two-state problems.

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Key words: cold molecule formation, Demkov–Kunike model, strong interaction limit

1. INTRODUCTION

During the last decade, creation of ultra-cold molecular systems has been of vast interest due to the potential for improved understanding of molecular physics and interactions, exploring new types of many-body systems such as condensates of dipolar molecules [1, 2], the BCS-BEC crossover [3], and the generation of entangled atoms by controlled dissociation of molecules [4]. To create ultracold molecules starting from laser-cooled atomic gases, two basic techniques, the Raman photoassociation [5] and magnetic Feshbach resonance [6], have been developed. Several experimental groups succeeded in the conversion of a macroscopic number of quantum-degenerate atoms into diatomic molecules starting from either a Bose–Einstein condensate [7] or a degenerate Fermi gas [8]. The highlight of these efforts was the observation of a molecular Bose–Einstein condensate [9].

In the present study we investigate the temporal dynamics of coherent molecule formation via photo- or Feshbach-association of ultracold atoms. We assume that all the atoms and molecules existing in the system belong to condensates of zero-momentum atoms and zero-momentum molecules, respectively. In this case, within the mean-field approximation, the coherent conversion of bosonic atoms into diatomic molecules (via either photo- or Feshbach-association) is governed by the following set of coupled nonlinear equations [10, 11]:

$$\begin{aligned} i\frac{da_1}{dt} &= U(t)e^{-i\delta(t)}\overline{a}_1 a_2, \\ i\frac{da_2}{dt} &= \frac{U(t)}{2}e^{i\delta(t)}a_1 a_1, \end{aligned} \tag{1}$$

where t is time, a_1 and a_2 will be referred to as the atomic and molecular state probability amplitudes, respectively, $\overline{a}_1$ denotes the complex conjugate of a_1. The real function $\delta(t)$ is defined as the integral of the detuning δ_t (the subscript denotes the derivative with respect to time) while the detuning δ_t defines the difference in energy $\hbar\delta_t$ between a zero-momentum molecule and two zero-momentum atoms which can be adjusted by tuning the laser field frequency in the case of photoassociation or by variation of the magnetic field in the case of Feshbach-association. In the case of photoassociation the atom–molecule coupling U can be controlled by variation of the laser field intensity, while in the case of Feshbach

resonance it is a fixed constant. System (1) describes a lossless process, i.e., it preserves the total number of particles that we normalize to unity: $|a_1|^2 + 2|a_2|^2 = \text{const} = 1$. In the present paper we consider the basic situation when the system starts from the all-atomic state: $|a_1(-\infty)| = 1,\ a_2(-\infty) = 0$.

So far, the major part of the theoretical developments to describe the cold molecule formation has been devoted to the case when the external field configuration is defined by the constant-amplitude linear level-crossing Landau–Zener (LZ) model [12]:

$$U = U_0, \quad \delta_t = 2\delta_0 t \tag{2}$$

(see, e.g., [13–25]). Consequently, when suggesting a qualitative interpretation of the observed experimental data, the intuition based on the analysis of the LZ model has typically been employed. However, it has been previously indicated that the simple intuition gained from the analysis of the LZ model becomes invalid when one considers, for instance, models with constant amplitude and finite final detuning [26]. In these cases, in contrast to the LZ prediction, the application of high laser field intensities (if the photoassociation terminology is used) is not always optimal for molecule formation. Hence, it is necessary to have a systematic knowledge how the variation of the coupling's and detuning's shapes affect the nonlinear dynamics of the system. To this end, in the present paper we analyze the quasi-linear level-crossing Demkov–Kunike model [27], characterized by a bell-shaped coupling and a finite variation of the detuning (see Fig. 1):

$$U = U_0 \operatorname{sech}\left(\frac{t}{\tau}\right), \quad \delta_t = 2\delta_0 \tanh\left(\frac{t}{\tau}\right) \quad (\tau > 0), \tag{3}$$

where τ is a (positive) scaling parameter. Without loss of generality we put $\tau = 1$ in what follows.

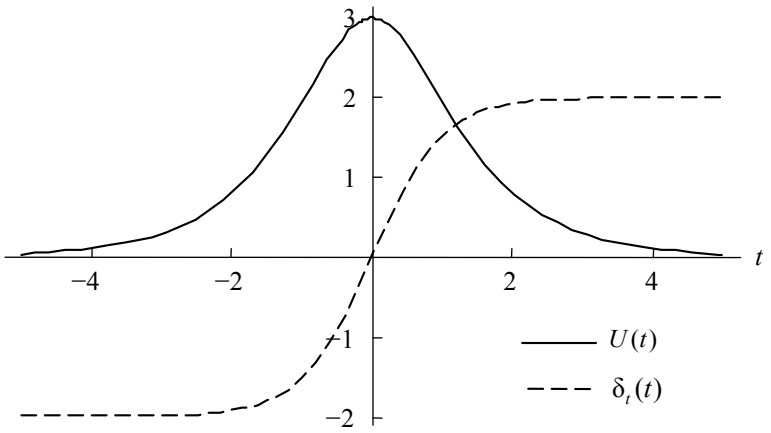


Fig. 1. The DK model: $U(t) = U_0 \operatorname{sech}(t),\ \ \delta_t(t) = 2\delta_0 \tanh(t)$.

The weak interaction regime of the nonlinear DK problem, corresponding to the small values of the peak coupling ($U_0 << 1,\ \forall\delta_0$) and very fast sweeps through the resonance in the case of the large peak coupling ($U_0 > 1,\ \delta_0 >> U_0$), has already been discussed in [28, 29]. In particular, in [29], an approximation describing the temporal dynamics of the molecule formation in the weak interaction regime has been presented. The strong interaction regime of the DK model, corresponding to the large values of the peak coupling U_0 and moderate values of the sweep rate through the resonance ($U_0 > 1$, $\delta_0 \le U_0$) has been studied in [30, 31], where two distinct scenarios of evolution are shown to be available – the small and large sweep rate regimes (see Fig. 2). The main peculiarity of the molecule formation process in the slow resonance sweep regime ($U_0 > 1,\ \delta_0 < 1$) is its pronounced oscillatory behavior: the system displays large-amplitude Rabi-type oscillations between the atomic and molecular populations long before the resonance crossing. On the contrary, in the fast resonance sweep regime of the strong interaction limit ($U_0 > 1,\ 1 \le \delta_0 \le U_0$) only weak, damped oscillations between the atomic and molecular subsystems occur; for the all-atom initial condition at $t \to -\infty$ the oscillations start up only after the resonance crossing time-point has passed and gradually vanish in time. An approximate solution describing the molecule formation in this limit has also been constructed in [30, 31]. However, the constructed approximation fails to predict the number of the molecules formed at the end of the interaction ($t \to +\infty$) and has a derivative discontinuity.

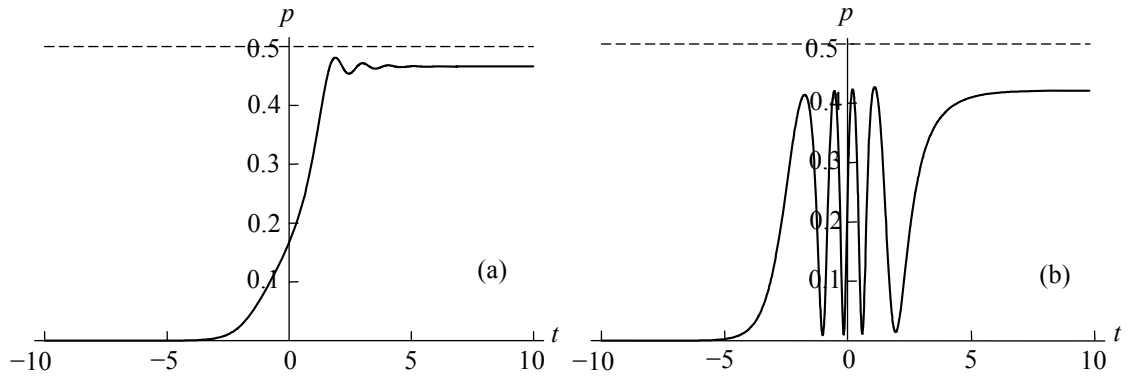


Fig. 2. The probability of the molecular state versus time for the DK model: (a) the fast resonance sweep regime of the strong interaction limit ($U_0 = 8,\ \ \delta_0 = 8$); (b) the slow resonance sweep regime of the strong interaction limit ($U_0 = 8,\ \delta_0 = 0.1$).

In the present paper we construct an approximate solution to the problem to describe the temporal dynamics of the molecule formation in the large sweep rate regime of the strong interaction limit. In contrast to the approximate solution constructed in [30, 31], the approximation presented in the present paper is a smooth bounded step-wise function which contains an adjustable fitting parameter. Due to these properties, one can use the presented solution as a zero-order approximation and construct the next approximation term to the problem. Importantly, the suggested approach can be interpreted as a modification of the adiabatic approximation [22–25] and can further be used for the treatment of analogous nonlinear two-state problems.

2. MATHEMATICAL TREATMENT

Based on our previous experience [29–31], we first reduce the governing set of equations (1) to a constant-amplitude form by applying the transformation of the independent variable

$$z(t) = \int_0^t \frac{U(t')}{U_0} dt'. \tag{4}$$

As it has been mentioned above, in the case of the Feshbach-association the coupling U is constant. Hence, in its direct form the DK model defined by Eq. (3) can only be used for the description of the photoassociation process. However, due to the transformation (4) the DK model is equally applicable to the case of the Feshbach-association.

Further, we write an exact equation obeyed by the function $p = |a_2|^2$:

$$p_{zzz} - \frac{\delta^*_{zz}}{\delta^*_z} p_{zz} + \left[\delta^{*2}_z + 4\lambda(1-3p)\right] p_z + \frac{\lambda}{2}\frac{\delta^*_{zz}}{\delta^*_z}\left(1 - 8p + 12p^2\right) = 0, \tag{5}$$

where the effective detuning δ^*_z is defined as

$$\delta^*_z(z(t)) = \delta_t(t)\frac{\sqrt{\lambda}}{U(t)} \tag{6}$$

with $\lambda = U_0^2$. It can easily be seen that in the case of the DK model $\delta^*_z(z(t)) = 2\delta_0 \sinh(t)$. Hereafter we conventionally refer to $p = |a_2|^2$ as to the molecular state probability (note that $p \in [0, 1/2]$).

As it has been mentioned above, in the large sweep rate regime of the strong interaction limit the parameters λ and δ_0 are supposed to be large, and the solution of Eq. (5) is almost non-oscillatory in this regime (see Fig. 2a). Thus, following [30, 31], we make an attempt to construct a valid approximation $p_0(t)$ by neglecting the two higher order derivative terms in the exact equation (5):

$$\left[\delta^{*2}_z + 4\lambda(1-3p_0)\right] p_{0z} + \frac{\lambda}{2}\frac{\delta^*_{zz}}{\delta^*_z}\left(1 - 8p_0 + 12p_0^2\right) = 0. \tag{7}$$

It can immediately be seen that this equation allows two trivial stationary solutions, $p_0 = 1/2$ and $p_0 = 1/6$.

In spite of its complexity, the limit equation (7) is exactly solvable (for a detailed derivation see [30, 31]). The general solution of this equation can be constructed by application of the transformation of the independent variable $s = \lambda/\delta_z^{*2}$ with further interchange of the dependent and independent variables. By applying this procedure, one arrives at a linear equation for s which is readily integrated, resulting in the following quartic polynomial equation for p_0:

$$\frac{\lambda}{\delta_z^{*2}} = \frac{C_0 + p_0(p_0 - 1/2)^2}{9(p_0 - 1/6)^2(p_0 - 1/2)^2}, \tag{8}$$

where C_0 is the integration constant. Now, in order to meet the initial condition $p_0(-\infty) = 0$ we put $C_0 = 0$. The quartic equation (8) is then reduced to a quadratic one, and we should choose the appropriate root of this equation that satisfies also the mentioned initial conditions. Taking into account Eqs. (6) and (8), we arrive at the following approximate solution for the nonlinear DK problem:

$$p_0(t) = \frac{1}{6} + \frac{\sinh(t)\left(\sinh(t) + \sqrt{\sinh^2(t) + 6\varepsilon}\right)}{18\varepsilon}, \tag{9}$$

where $\varepsilon = \lambda/4\delta_0^{\,2}$. However, for $t > \operatorname{arcsinh}(\sqrt{2\varepsilon})$, $p_0(t)$ defined by this formula exceeds $1/2$ allowed by the normalization. Moreover, the solution (9) is not bounded when $t \to +\infty$. In [30, 31] it has been suggested to overcome this inconsistency via combination of the function (9) with the trivial solution $p_0 = 1/2$.

Though the approximation constructed in this way quite well describes the association process before the resonance crossing and not long after the crossing, it suffers from substantial shortcomings: it does not predict the correct value for the final transition probability and it has a derivative discontinuity at the point $t = t_0$. However, as it will be shown below, it is possible to modify the limit equation (7) in such a way that it will have a bounded stepwise solution. We first note that if one takes a nonzero value for the integration constant C_0 then, generally speaking, the quartic equation (8) does not reduce to a quadratic one, and the function $p_0(t)$ does not diverge at $t \to +\infty$. But for nonzero C_0 the constructed approximate solution does not satisfy the initial condition $p_0(-\infty) = 0$. This observation gives a hint that when constructing an approximation one should try to avoid the degeneracy of the quartic polynomial equation to a quadratic one. It is possible to resolve this issue via an appropriate modification of the limit equation (7) by introducing into it a term of the form $A\delta_{zz}^*/\delta_z^*$, where A is an adjustable parameter. In this way, we arrive at the following augmented limit equation:

$$\left[\delta_z^{*2} + 4\lambda(1 - 3p_0)\right]p_{0z} + \frac{\lambda}{2}\frac{\delta_{zz}^*}{\delta_z^*}\left(1 - 8p_0 + 12p_0^2\right) - A\frac{\delta_{zz}^*}{\delta_z^*} = 0, \tag{10}$$

which can be integrated using the method applied for solving equation (8). The integration leads to a remarkable result:

$$\frac{\lambda}{\delta_z^{*2}(z(t))} = \frac{p_0(p_0 - \beta_1)(p_0 - \beta_2) + C_0}{9(p_0 - \alpha_1)^2(p_0 - \alpha_2)^2}, \tag{11}$$

where

$$\alpha_{1,2} = \frac{1}{3} \mp \frac{1}{6}\sqrt{1 + \frac{6A}{\lambda}}, \quad \beta_{1,2} = \frac{1}{2} \mp \sqrt{\frac{A}{2\lambda}}. \tag{12}$$

This relation defines a quartic algebraic equation for determination of the function $p_0(t)$. The determination of the fitting parameter A will be discussed later.

Now we analyze in detail the solution of the quartic equation (11) for the case of the DK model. First of all, we note that for the initial condition $p_0(-\infty) = 0$, it holds $C_0 = 0$. Then we note that at $t \to +\infty$ the left-hand side of Eq. (11) tends to zero. Hence, either $p_0(+\infty) = \beta_1$ or β_2. Since $\beta_2 > 1/2$, we conclude that $p_0(+\infty) = \beta_1$. Furthermore, considering the behavior of p_0 at $t = 0$ we see that at $t \to 0$ the left-hand side of Eq. (11) diverges, hence, $p_0(0) = \alpha_1$ because $\alpha_2 > 1/2$. Summarizing the results, we have

$$p_0(0) = \frac{1}{3} - \frac{1}{6}\sqrt{1 + \frac{6A}{\lambda}}, \quad p_0(+\infty) = \frac{1}{2} - \sqrt{\frac{A}{2\lambda}}. \tag{13}$$

Note that $p_0(0) < 1/6$ and $p_0(+\infty) < 1/2$. The limit solution $p_0(z(t), A)$ is a monotonically increasing function that starts from zero at $t = -\infty$, reaches some value less than $1/6$ at $t = 0$ and tends to a finite positive value less than $1/2$ for $t \to +\infty$ when $0 < A < \lambda/2$ (see Fig. 3).

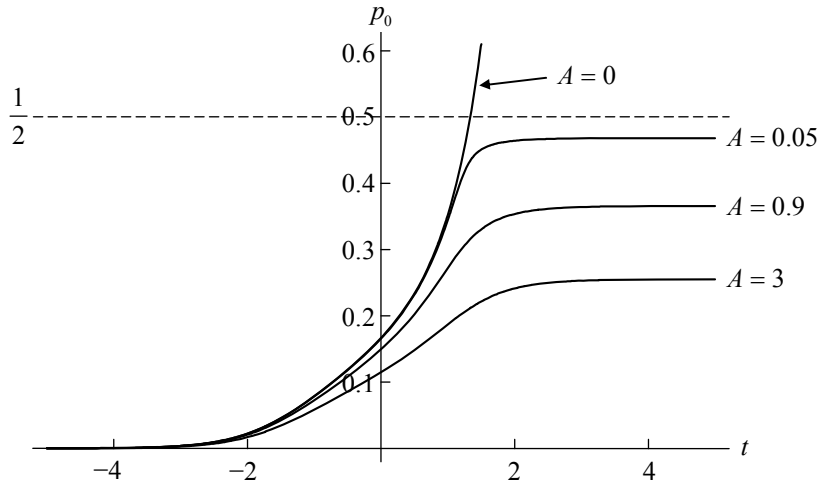


Fig. 3. The limit solution $p_0(t)$ (see Eq. (11)) versus time at $\lambda = 25$ for $A = 0$, 0.05, 0.9, and 3.

To develop general principles from which the fitting parameter A could be determined, we insert the approximate solution $p_0(z, A)$ into the exact equation for the molecular state probability (5) and consider the behavior of the remainder

$$R = \left(\frac{d}{dz} - \frac{\delta^*_{zz}}{\delta^*_z} \right) \left(p_{0zz} - A \right). \tag{14}$$

It is intuitively understood that the better approximation p_0 is, the smaller remainder R will become (it would be identically zero if p_0 were the exact solution to Eq. (5)). Thus, we try to minimize the remainder via an appropriate choice of the fitting parameter A. The remainder R is not bounded at the resonance point $z = 0$. To eliminate the divergence of the remainder at this point of singularity, we choose the parameter A as a solution of the equation

$$p_{0zz}(0) - A = 0. \tag{15}$$

Taking into account the definition of p_0, i.e., Eqs. (11) and (12), in the case of the strong coupling ($\lambda >> 1$) and moderate values of the sweep rate δ_0 ($1 < \delta_0 < \sqrt{\lambda}$) we obtain the following approximate expression for the parameter A:

$$A \approx \frac{\lambda}{27 \delta_0^{\sqrt{3}}}. \tag{16}$$

It should be noted that a more accurate approximation for A valid for a larger region of variation of δ_0 and λ is of the form $A \approx \lambda / g(\delta_0)$ with a complicated function $g(\delta_0)$ vanishing at $\delta_0 \to \infty$. From the qualitative point of view, however, the approximation (16) reveals the important peculiarity that we would like to note here, namely, the linear dependence of A on λ in the strong coupling limit.

It should also be noted that the limit solution p_0 still misses several essential features of the association process. Indeed, for instance, the coherent oscillations between atomic and molecular populations which come up after the system has passed through the resonance are not contained in this solution (see Figs. 2a and 3). Furthermore, the final transition probability at $t \to +\infty$ predicted by p_0, with the parameter A defined by Eq. (16), is always lower than what is shown by the numerical solution of the exact equation.

A natural conclusion is that the shortcomings of the suggested limit solution are due to the singular procedure we have employed to derive it. Indeed, we have constructed p_0 by neglecting the two higher order derivative terms in Eq. (5). Of course, these terms played a role in determination of the appropriate value of A, hence, they have been taken into account, to some extent. However, for a considerable

improvement of the obtained result, we need the next correction term that counts for the second and third order derivatives of p. Importantly, one can use the presented solution p_0 as a zero-order approximation to construct the next approximation term to the problem.

3. CONCLUSION

We have studied the nonlinear dynamics of ultracold molecule formation via coherent photo- and Feshbach-association of ultracold atoms for the case when the external field configuration is defined by the quasi-linear level-crossing DK model (3), characterized by a bell-shaped coupling and finite variation of the detuning. In the present paper we have focused on the large sweep rate regime of the strong interaction limit ($\lambda > 1,\ \ 1 < \delta_0 < \sqrt{\lambda}$). In the case of photoassociation the discussed regime models a situation when the peak laser field amplitude is quite large, and the laser, being far detuned in the beginning of the interaction passes, linearly in time, through the resonance, and eventually becomes again far detuned (see Eq. (3)).

To describe this regime quantitatively, we have analyzed the equation for the molecular state probability (5). We have constructed a zero-order approximation to the problem which has been defined as a solution of the first-order nonlinear equation (10). We have shown that the exact solution of this equation is given as a solution of the polynomial equation of fourth order (see Eq. (11)). The constructed approximation contains a fitting parameter which has been determined through a variational procedure. Being a step-wise bounded smooth function (see Fig. 3), p_0 can be used as a zero-order approximation to construct the next approximation term to the problem. Note that the presented developments are not restricted to the case of the DK model only: they are equally applicable to other level-crossing models.

Finally, we would like to note that nevertheless the adiabatic theorem breaks down for the nonlinear two-state problem under consideration (1) [22–25], resulting in the divergence of the adiabatic approximation given by Eq. (8), we still succeeded in the construction of a valid approximation via modification of the limit equation (7). From this point of view the suggested approach can be interpreted as a general prescription to eliminate the divergence of the adiabatic approximation (8).

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