Controlled interference of association paths in the conversion of ultracold atoms into molecules

J Plata

Departamento de Física, Universidad de La Laguna, La Laguna E38204, Tenerife, Spain

E-mail: jmplata@ull.es

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Abstract

We present a proposal for controlling the conversion of ultracold atoms into molecules by fixing the phase difference between two oscillating magnetic fields. The scheme is based on the use of a magnetic Feshbach resonance with a field modulation that incorporates terms oscillating with frequencies corresponding to the main resonance and one of the subharmonics. The interference between the two association processes activated by the oscillating terms is controlled via the phase difference. As a result, significant increase or decrease of the effective interaction strength can be achieved. The realization of the proposal is feasible under standard technical conditions. In particular, the method is found to be robust against the effect of the sources of decoherence present in the practical setup. The applicability of the approach to deal with quadratic terms in the field modulation is discussed.

Keywords: ultracold atoms and molecules, Feshbach resonance, methods of control

1. Introduction

The use of Feshbach resonances (FRs) has been crucial for advances made in recent years in the study of ultracold atoms and molecules [1–9]. The control of the position and width of a FR, and consequently, the manipulation of the dynamics of the atom-molecule system, have been the objectives of intense research. In this line, interesting proposals, which incorporate the use of different fields and transitions, have been made [10–13]. These control issues are particularly relevant to the optimization of the methods employed in the production of ultracold molecules. (The progress in this area is continuous; in this sense, it is worth pointing out interesting experimental work reported in [14–17].) One of the techniques frequently applied to generate ultracold molecules from both Fermi and Bose atomic gases is based on a sinusoidal modulation of the field in a magnetic FR [18–23]. With this arrangement, molecule formation has been observed not only at the main resonance, i.e., for a field frequency matching the molecular binding energy, but also at subharmonic resonances. Here, we propose a variation of this scheme that introduces additional elements of control in the conversion process. Specifically, we consider a magnetic modulation that incorporates two terms oscillating with different frequencies. In the particular case proposed to illustrate the method, one of the frequencies corresponds to the main resonance and the other to one of the subharmonics. The phase difference between the driving terms can be used to steer the interference between the processes generated via the two considered resonances, and, in turn, to optimize (or inhibit) the production of molecules. Our approach to the system, based on the use of appropriate unitary transformations to simplify the description, allows an analytical characterization of the transition process, and, consequently, a direct application of the results to the implementation of strategies of control. The robustness of the scheme against the dephasing mechanisms potentially present in the considered scenario is evaluated. The practical realization of the interference paths, viable under usual experimental conditions, can serve to assess our understanding of the subharmonics resonances. It can also provide a valuable test on the assumed coherent response of the atom-molecule system. The general components of the developed method, combined in different forms, can found applicability in various contexts. In this sense, differential implications of variations of the basic scheme are discussed. The outline of this paper is as follows. In section 2, a first general description of basic two-body dynamical characteristics...
of the system is presented. The specific proposal for the interference scheme is introduced in section 3. Additionally, the applicability of the theoretical framework to account for second-order effects in the modulation field is assessed. In section 4, we analyze many-body aspects of the dynamics and discuss the relevance of the different sources of decoherence present in the system realization to the validity of the method. Finally, some general conclusions are summarized in section 5.

2. Two-body dynamics

We consider a Bose gas of ultracold atoms in a weakly confining harmonic trap. In order to induce molecule association, a magnetic FR is applied: the variation of the magnetic field alters the detuning between the (closed-channel) FR state $|R\rangle$ and the (entrance-channel) atom state $|S\rangle$, and, consequently, modifies the effectiveness of the inter-channel coupling. This opens the possibility of manipulating the scattering length. Moreover, depending on its functional form, the time-dependent field can induce transitions between the entrance states and the bound state. This second implication will be crucial for the scheme proposed in our study. (As in the original FR scenario, we consider that the interchannel coupling is due to hyperfine interactions; the generalization of our approach to deal with modifications of the coupling is direct.) Hence, in the considered arrangement, the magnetic field is modulated as

$$B(t) = B_0 + B_m(t),$$

where the average value $B_0$ is assumed to be close to a FR position $B_s$. Additionally, in order to work in a perturbative regime, we consider that the modulation field $B_m$ is sufficiently small, $|B_m| \ll |B_0 - B_s|$. (The proposed functional form of $B_m(t)$, appropriate for our strategies of control, will be specified further on.) The weak-confinement conditions allow us to apply a local-density approximation. In our procedure, we focus on the simplest case of a uniform system. Consistently, the atomic energy levels, which are closely spaced because of the weak trapping, will be approximated as a quasi-continuum. The robustness of the approach against the effect of nonuniformities in the field will be discussed.

2.1. The undriven system: the bare and the dressed-state representations

In the absence of magnetic modulation, (i.e. for $B_m = 0$), the Hamiltonian reads $H_1 = H_0 + V$, where $H_0$ stands for the Hamiltonian of the interaction-less system at the mean magnetic field $B_0$, and $V$ represents the coupling term between the eigenstates of $H_0$, i.e., the bare states, $|R\rangle$ and $|S\rangle$. Specifically, we have [22]

$$H_0|S\rangle = E_S|S\rangle, \quad 0 \leq E_S < \infty,$n$$

$$H_0|R\rangle = -\epsilon_{B_0} |R\rangle, \quad \epsilon_{B_0} > 0,$n$$

$$\langle R | V | S \rangle = v(E_S),$$

where $\epsilon_{B_0}$ denotes the binding energy of the state $|R\rangle$ at the field $B_0$, and $E_S = p^2/m$ represents the energy along the atomic set, $p$ is the relative linear momentum and $m$ stands for the atom mass.) The operator $V$, which accounts for hyperfine interaction between the atom pairs in the open and in the closed channels, affects only nuclear and electron spin variables, which are fixed in each channel. Therefore, the dependence of the interaction strength $v(E_S)$ on the atom-state characteristics is determined by the overlap between the wave functions $\psi_R(\vec{r}) = \langle \vec{r} | R \rangle$ and $\psi_S^{(p)}(\vec{r}) = \langle \vec{r} | S \rangle$, where $\vec{r}$ denotes the relative coordinate. Then, the functional form of $v(E_S)$ can be determined through the evaluation of the Franck-Condon factor

$$F(p) \equiv \int d\vec{r} \overline{\psi_R(\vec{r})\psi_S^{(p)}(\vec{r})}^2.$$

In the considered range of energies, $v(E_S)$ can be assumed to hardly vary with $E_S$. Moreover, we assume that the set of dressed states of the system, (i.e., of eigenstates of the complete Hamiltonian $H_1$), is formed by a bound eigenstate $|M\rangle$, (the molecular state), with energy $-\epsilon_{M}$, and by a continuum of states $|A\rangle$ with energy $E_A$. Namely,

$$H_1 |A\rangle = E_A |A\rangle, \quad 0 \leq E_A < \infty,$n$$

$$H_1 |M\rangle = -\epsilon_{M} |M\rangle, \quad \epsilon_{M} > 0.$$

As the field value $B_0$ approaches the FR position $B_s$, the dressed and the bare-state representations increasingly differ. In particular, $|M\rangle$ departs from $|R\rangle$. Hence, in the considered situations, which require the description of atom-molecule transfer processes near $B_s$, the use of the dressed-state basis is necessary.

2.2. The driven dynamics in the dressed-state representation

We assume that, as in standard practical setups [18], the time-dependent magnetic field is applied through a trapezoidal ramp similar to that outlined in figure 1. In the first linear segment, the field is slowly varied from $B(t = t_1)$ (far from the FR), to $B_0$. In the central plateau, the modulation $B_m(t)$ introduced in equation (1) is connected. Subsequently, a slow linear ramp drives $B(t)$ back to the initial value. A detailed study of the incorporation of this type of arrangements in the theoretical description was presented in [22].

Figure 1. A diagram (with arbitrary scales) of the magnetic-field ramp.
methodology applied there combines the following elements. First, the effects of the (slow) linear segments of the ramp are simulated through an adiabatic approximation that connects the bare and dressed representations. Second, to deal with the dynamics in the central region, a dressed-state approach is implemented. The adiabatic segments can be regarded as part of the system preparation. (Their role in different practical setups, i.e. in arrangements corresponding to a condensate preparation and to an initial thermal mixture, was analyzed in [22].) Since, that analysis is applicable to the present context, we will concentrate here on the differential aspects introduced by the considered modulation in the dynamics in the central plateau.

The incorporation of the magnetic modulation into our theoretical framework is straightforward in the bare-state picture: the field variation alters the binding energy of the FR state according to \( \epsilon_B \rightarrow \epsilon_B + \hbar C_B B_m(t) \), where \( \hbar C_B = \left[ \frac{\partial}{\partial B} \right]_{B_0} \) corresponds to the difference between the magnetic moments of the involved states. (In the following, we will take \( \hbar = 1 \). The effect of field-quadratic corrections to the binding energy will be considered later on.) As a consequence, the complete system is described by the Hamiltonian \( H_2 = H_1 + C_B B_m(t) |R \rangle \langle R| \), which, in the dressed-state basis, has the form

\[
H_2 = \left[ - \epsilon_M + \eta_B B_m(t) \right] |M \rangle \langle M| + E_\alpha(A) |A \rangle \langle A| + \left[ \eta_M B_m(t) |M \rangle \langle A| + \text{h.c.} \right],
\]

where

\[
\eta_M = C_{B_0} |\langle M | R \rangle|^2
\]

\[
\eta_\alpha(A) = C_{B_0} |\langle A | R \rangle|^2
\]

\[
\text{and}
\]

\[
\eta_M = C_{B_0} |\langle M | R \rangle| \langle A| R \rangle.
\]

Notice that the field modulation has a twofold effect: it introduces a time-dependent coupling between the states \( |M \rangle \) and \( |A \rangle \), and induces a time variation in their eigenenergies. Although the modulation can also couple different dressed atomic states, that interaction has been neglected due to the small magnitude of the involved state-projections, and given that, in the considered situations the atomic energy spacing is far from being in resonance with the driving field.

3. The interference scheme

3.1. General approach

Our proposal for manipulating the dynamics is based on the application of a magnetic-field modulation \( B_m(t) \) composed of two oscillating terms with different frequencies and amplitudes and a constant phase difference. (The generalization to more than two terms is direct.) Namely, in the central region of the previously indicated trapezoidal ramp, we connect a modulation field with the form

\[
B_m(t) = B_1 \sin(\omega_1 t) + B_2 \sin(\omega_2 t + \chi).
\]

To tackle the resulting dynamics, we first apply the unitary transformation

\[
U(t) = e^{i \left[ (\omega t + \eta_B F(t)) |M \rangle \langle M| + \eta_M F(t) |A \rangle \langle A| \right]}
\]

where we have introduced the function

\[
F(t) = \frac{B_1}{\omega_1} \cos(\omega_1 t) + \frac{B_2}{\omega_2} \cos(\omega_2 t + \chi).
\]

(The value of \( \omega \) will be chosen, depending on the particular (frequency) characteristics of the scheme, to simplify the picture brought about by the application of \( U(t) \). As it corresponds to a time-dependent unitary transformation [24], the transformed Hamiltonian is given by

\[
H_2' = U' H_2 U - i U' U.\]

Now, for the terms that contain \( F(t) \), we take into account the expansion of the exponential of a sine or cosine function in terms of the Bessel functions \( J_k(x) \) [25], and rewrite the Hamiltonian as

\[
H_2' = ( - \epsilon_M + \omega ) |M \rangle \langle M| + E_\alpha(A) |A \rangle \langle A| + \text{h.c.} + \left[ \eta_M B_m(t) |M \rangle \langle A| + \text{h.c.} \right].
\]

where we have incorporated the functions

\[
G_j(t) = \left\{ \begin{array}{ll}
\frac{1}{2} \sum_{m=-\infty}^{\infty} \sum_{q=-\infty}^{\infty} (-1)^{m+q+j} \left( \zeta_1 B_1 \right) \left( \zeta_j B_j \right) \\
\times e^{i[(\omega_m+\omega_{q+j}+q\omega_j)t+q\chi]} (e^{i\omega_j t} - e^{-i\omega_j t}) \end{array} \right\}, \quad (j = 1, 2),
\]

and the coefficients

\[
\zeta_j = \frac{\eta_M - \eta_\alpha}{\omega_j}, \quad (j = 1, 2).
\]

The form of \( H_2' \) given by equation (13) along with equations (14) and (15) provides a framework for simplifying the description. Specifically, an analytical coarse-grained picture of the dynamics can be derived through the averaging of \( H_2' \). Apart from the diagonal term, (where the effective detuning \( - \epsilon_M + \omega \) will be minimized by the appropriate choice of \( \omega \)), \( H_2' \) has a series of interaction terms oscillating with frequencies determined by \( \omega_1, \omega_2, \omega_j \), and by the integer numbers \( m \) and \( q \). The secular dynamics is then governed by the components with zero frequencies. The rest of terms barely affect the system evolution: they present fast oscillations, which, because of the small magnitude of their amplitudes compared with the involved frequencies, can be averaged out to zero. (We recall that a perturbative regime is considered for the modulation amplitudes.) The secular terms can be easily identified if \( \omega \) is appropriately chosen as a function of the characteristic frequencies of the driving. Additionally, for the previous arguments on the negligible role of the highly oscillating terms to be applicable, the choice of \( \omega \) must lead to a significant reduction of the effective detuning. This general procedure, which is applicable to
different combinations of frequencies and amplitudes of the driving field, will be particularized, in the following, to a specific proposal.

3.2. Interference of the association paths corresponding to the main resonance and the first subharmonic

Let us consider a particular arrangement where the frequencies of the driving field are \( \omega_1 = 2\omega_2 \approx \epsilon_M \). Then, the appropriate value of the characteristic frequency \( \omega_c \) to be introduced in equation (11), is \( \omega = \omega_1 \); this choice minimizes the effective detuning and allows us to straightforwardly single out the secular terms in equation (14). We can go further by identifying the dominant contribution among those terms. Indeed, taking into account the perturbative character of the modulation field, the arguments of the Bessel functions in equation (14) can be assumed to be small enough for the magnitude of those functions to significantly decrease as the absolute value of the integer index grows. Hence, the dominant stationary term in \( G_{\ell}(t) \) corresponds to the indexes \( m = 0, q = 0 \). In parallel, in \( G_{2\ell}(t) \), the most important secular contribution comes from the term with \( m = 0, q = -1 \). (In order to simplify the presentation, we focus on the effect of the first-order contributions. The inclusion of the higher-order corrections, albeit lengthy, is direct.) Therefore, in the optimum frequency range, namely, for \( \omega = \omega_1 \sim \epsilon_M \), the dynamics is approximately described by the reduced Hamiltonian

\[
\hat{H}_{\text{eff}} = \hat{E}_M |M\rangle \langle M| + \hat{E}_A |A\rangle \langle A| + \left[ \tilde{v}_{\text{eff}} \left( E_A |M\rangle \langle A| + \text{h.c.} \right) \right],
\]

where the effective energy of the molecular state is given by \( \hat{E}_M = \omega_1 - \epsilon_M \), and

\[
\tilde{v}_{\text{eff}} \equiv -J_0(\zeta_1 B_1) \left[ iJ_0(\zeta_2 B_2) B_1 + J_1(\zeta_2 B_2) e^{i\chi} B_2 \right] \frac{v_{\text{eff}}}{2}
\]

is a renormalized coupling constant which incorporates the magnetic modulation. As the splitting between the effective atomic and molecular levels reads \( \Delta \equiv \hat{E}_M - \hat{E}_A = \omega_1 - (\epsilon_M + E_A) \), the resonance frequency is given by \( \omega_{\text{res}} = \epsilon_M + E_A \). From these results, some preliminary conclusions can be drawn:

(i) \( \tilde{v}_{\text{eff}} \) presents a nontrivial dependence on the field amplitudes: in addition to appearing as explicit factors in the above expression, \( B_1 \) and \( B_2 \) enter the arguments of the Bessel functions. Actually, because of the form of those functions, there is no monotonous increase of the effective interaction strength with the amplitudes of the field components. Moreover, outside the limited range of arguments where the applied perturbative treatment is valid, a complex dependence of the effective coupling term on \( B_1 \) and \( B_2 \) can be expected.

It is also worth stressing the relevance of the Franck-Condon factor \( F(p) \) (see equation (3)), to the derived effective strength. \( F(p) \), which determines the magnitude of the original coupling, and, consequently, the form of the dressed states, enters \( v_{\text{eff}} \) via the product of state projections \( \langle M| R \rangle \langle R| A \rangle \).

Figure 2. Magnitude of the effective interaction strength \( |v_{\text{eff}}| \) as a function of the phase difference \( \chi \) for two sets of magnetic-field amplitudes: \( B_1 = 0.255 \) and \( B_2 = 1.0 \) (a); \( B_1 = 0.5 \), and \( B_2 = 0.3 \) (b). In both cases, \( \zeta_1 = 1.0 \), and \( \zeta_2 = 0.5 \). (The phase difference is expressed in radians; arbitrary units are used for the rest of magnitudes.)

(ii) It is apparent that the conversion process can be controlled through the choice of the phase difference \( \chi \). Significant changes in the effective interaction strength can be induced by varying \( \chi \). In particular, by taking \( \chi = \pi/2(-\pi/2) \), a constructive (destructive) interference is brought about. Furthermore, for \( \chi = -\pi/2 \), a complete inhibition of the molecule generation process can be achieved by properly choosing the values of the field amplitudes. Figure 1 corresponds to a set of parameters that exemplifies that situation.

(iii) From equation (17), one consistently recovers the results obtained for a single-frequency driving field in [22]. Specifically, by taking \( B_2 = 0 \), the result found for the main resonance is reproduced. Alternatively, by fixing \( B_1 = 0 \), we obtain the renormalized coupling constant associated with the first subharmonic resonance. In our framework, it is possible to obtain the field amplitude \( B_1 \) (or \( B_2 \)) that optimizes the effective interaction strength \( v_{\text{eff}} \) for the main-resonance case (or for the first subharmonic). It is worth stressing the potential practical interest of the use of subharmonic frequencies: they can allow the access to binding energies that are outside the available bandwidth of the current techniques for generating magnetic fields.

(iv) The additional elements of control introduced in the molecule-association setup configure a more versatile scenario. In it, a variety of behaviors can be implemented, for instance, via sudden or adiabatic changes of the set of field parameters. Similarities with the typical arrangements for Ramsey spectroscopy [3, 26, 27] can be traced: different frequencies can be apparent in the evolution of the system by connecting (or disconnecting) the oscillating terms. (Here, it is worth mentioning the interest of parallel setups, like that studied in [28].)

The above arguments are illustrated in figures 2 and 3. The case (a) in figure 2 exemplifies how significant changes in the magnitude of the effective coupling constant can be achieved by varying the phase difference. The previously
mentioned suppression of the conversion process for 
\( \chi = -\pi/2 \) and its intensification for \( \chi = \pi/2 \) are apparent.
Case (b), which corresponds to a different set of field
amplitudes, represents a more smooth variation of \( \tilde{\nu}_{\text{eff}} \) with \( \chi \). As can be shown from equation (17), the changes induced
by varying \( \chi \) are more drastic as the magnitudes of the two
terms that contribute to \( \tilde{\nu}_{\text{eff}} \) approach. Figure 3 illustrates
the dependence of the interaction strength on \( B_2 \). The variety of
behaviors that can emerge depending on the system parameters
is again evident. Note that the eventual monotonous
increase of \( \tilde{\nu}_{\text{eff}} \) with \( B_2 \) corresponds to the regime where
the role of the second field is dominant. (Here, it is worth
recalling the need of working with relatively small arguments
of the Bessel functions to guarantee the applicability of the
approach.) We emphasize that equation (17) allows
significant predictive power on the system dynamics; actually,
the possibility of using our proposal to control the conversion
process is apparent.

We stress that, in the present work, we focus on a pri-
mary aspect of the dynamics that can be used to control
the efficiency of the atom-molecule conversion process. Namely,
we concentrate on the variation of the effective coupling
constant that can be achieved with the proposed arrangements
of magnetic fields. Additional aspects of the dynamics, which
can limit the conversion efficiency, were analyzed for the case
of single-frequency field in [22]. The conclusions of that
study are directly applicable to the present scenario.

3.3. Applicability of the general approach to the analysis
of field-quadratic terms in the binding energy of the FR state

By now we have considered a perturbative regime for the
modulation field. Correspondingly, we have neglected sec-
ond-order effects in the variation of the original detuning \( \epsilon_{B_0} \)
induced by \( B_m(t) \). However, since those effects have been
observed in some of the experimental realizations of the
system, it is worth evaluating here their implications to the
dynamics. In this sense, we must recall the results of [20],
which uncovered the existence of a shift in the molecular
energy proportional to the square amplitude of the modulation
field. (In that case, a single-frequency driving field was
applied.) Actually, that energy displacement was found to
significantly modify the resonance condition. We will see that
the approach presented above can be adapted to account for
effects rooted in terms quadratic in the field \( B_m(t) \). Moreover,
we will show that the combined effects of first and second-
order terms in \( B_m(t) \) can be regarded as a particular case of the
interference scheme previously developed. Convenient for the
simplicity of the presentation is to carry out the analysis with
a single-frequency driving field, (as corresponds to the
experiments of [20].) The generalization of the study to the
case of a biharmonic driving will be discussed.

Hence, we assume now that the magnetic-field modula-
tion modifies the undriven Hamiltonian in the form
\( H_1 \rightarrow H_2 = H_1 + \left[ C^{(1)}_{B_0} B_m(t) + C^{(2)}_{B_0} B_m(t)^2 \right] \left| R \right\rangle \left\langle R \right| \),
where \( C^{(1)}_{B_0} \equiv \left( \frac{\partial \epsilon}{\partial B_0} \right)_{B_0} \) corresponds to the coefficient previously
denoted as \( C_{B_0} \), and we have introduced \( C^{(2)}_{B_0} = \frac{1}{2} \left( \frac{\partial^2 \epsilon}{\partial B_0^2} \right)_{B_0} \) to
characterize the (additional) quadratic dependence of the
binding energy on the magnetic field. Moreover, we take
\( B_m(t) = B_1 \sin(\omega_1 t) \), and rewrite \( H_2 \) as
\[
H_2 = H_1 + \left[ \frac{1}{2} C^{(2)}_{B_0} B_1^2 + C^{(1)}_{B_0} B_1 \sin(\omega_1 t) \right]
+ \frac{1}{2} C^{(2)}_{B_0} B_1^2 \sin(2\omega_1 t - \pi/2) \left| R \right\rangle \left\langle R \right|,
\]
which, in the basis of dressed states, (i.e., of eigenstates
of \( H_1 \)), is cast into the form given by equation (5). Namely, it is
expressed as
\[
H_2 = \left[ -\hat{\epsilon}_M + \eta_M \hat{B}_m(t) \right] |M\rangle \langle M|
+ \left[ \hat{E}_A + \eta_A (E_A) \hat{B}_m(t) \right] |A\rangle \langle A|
+ \left[ \nu_{\text{eff}} (E_A) \hat{B}_m(t) |M\rangle \langle A| + \text{h.c.} \right].
\] (18)
where we have introduced the displaced energies \( \hat{\epsilon}_M \) and \( \hat{E}_A \) given by
\[
\hat{\epsilon}_M = \epsilon_M - \eta_M \frac{1}{2} \frac{C^{(2)}_{B_0}}{C^{(1)}_{B_0}} B_1^2,
\] (19)
\[
\hat{E}_A = E_A + \eta_A (E_A) \frac{1}{2} \frac{C^{(2)}_{B_0}}{C^{(1)}_{B_0}} B_1^2,
\] (20)
and the effective biharmonic modulation field \( \hat{B}_m(t) \) defined as
\[
\hat{B}_m(t) = B_1 \sin(\omega_1 t) + \frac{1}{2} \frac{C^{(2)}_{B_0}}{C^{(1)}_{B_0}} B_1^2 \sin(2\omega_1 t - \pi/2).
\] (21)
Moreover, \( \eta_M, \eta_A (E_A) \), and \( \nu_{\text{eff}} (E_A) \) are respectively
given by equations (6), (7) and (8) where we only make the change
of notation \( C_{B_0} \rightarrow C^{(1)}_{B_0} \).
It is then apparent that the general approach introduced to
deal with the interference of association paths can be applied
to analyze the effect of incorporating field-quadratic terms into the bare-state detuning. Notice that the effective field \( \hat{B}_m(t) \) corresponds to a particular case of the generic biharmonic field given by equation (9) with \( B_2 = \frac{1}{2} \frac{C_2}{\hbar \omega_0} B_0^2 \), \( \omega_2 = 2\omega_1 \), and \( \chi = -\pi/2 \). Now, following the procedure introduced in the general approach, we apply the appropriate unitary transformation (with \( \omega = \omega_1 \)), [see equation (10)], and identify the dominant secular terms in the transformed Hamiltonian. Accordingly, the reduced Hamiltonian is found to be given by

\[
\hat{H}_{\text{eff}} = \hat{E}_M |M\rangle \langle M| + \hat{E}_A |A\rangle \langle A| + \left[ \hat{v}_{\text{eff}} (\hat{E}_A) |M\rangle \langle A| + \text{h.c.} \right],
\]

(22)

where \( \hat{E}_M = \omega_1 - \hat{\epsilon}_M \), and

\[
\hat{v}_{\text{eff}} = J_0(\zeta_1 B_1) \left[ -iJ_0(\zeta_1 B_1) B_1 + J_1(\zeta_1 B_1) B_2 \right] \frac{V_{\text{eff}}}{2}.
\]

Taking into account the expressions of the Bessel functions in the limit of small arguments and keeping only terms till second order in \( B_1 \), we can make the additional approximation for the effective interaction strength

\[
\hat{v}_{\text{eff}} = -iJ_0(\zeta_1 B_1) B_1 \frac{V_{\text{eff}}}{2}.
\]

This expression corresponds to that obtained when only linear terms are considered. Hence, at the considered level of approximation, the quadratic corrections affect only the splitting between the effective atomic and molecular levels, which now reads \( \hat{\Delta} \equiv \hat{E}_M - \hat{E}_A = \Delta - \left( \omega_M - \omega_A \right) \frac{1}{2} \frac{C_2}{\hbar \omega_0} B_1^2 \).

Consequently, the resonance frequency is given by

\[
\omega_1^{(R)} = \epsilon_M + E_A + \left( \omega_M - \omega_A \right) \frac{1}{2} \frac{C_2}{\hbar \omega_0} B_1^2.
\]

In agreement with the findings of [20], we have obtained a displacement of the resonance which is quadratic with the field amplitude. The magnitude of this shift depends on the (linear and quadratic) coefficients that characterize the field dependence of the detuning, and also on the projections between the FR state \( |R\rangle \) and the dressed states \( |M\rangle \) and \( |A\rangle \) involved in the transition.

It is apparent that in the case of considering a biharmonic function for \( \hat{B}_m(t) \), the quadratic shift of the resonance is still present. Since additional oscillating terms appear in the effective field \( \hat{B}_m(t) \), a more complex expression is obtained for \( \hat{v}_{\text{eff}} \).

4. Discussion of the relevance of the different sources of decoherence

The feasibility of controlling the interference between association paths depends crucially on the coherent response of the system to the modulation field. In the above description of the scheme, we deal with a simplified scenario where coherence was assumed. Now, we turn to analyze the robustness of the proposal against different components of the system which can affect the coherent character of the dynamics.

First, we recall that the atomic state involved in the transition has been assumed to be effectively isolated from the rest of the atomic set. Accordingly, equation (11) defines an effective (linear) Rabi model involving \( |M\rangle \) and an (isolated) generic state \( |A\rangle \). The quasi-continuum atomic structure, rooted in the confinement, can be incorporated into the model by describing the atomic set in terms of the density of states and of the coupling function with the bound state. Different positions of \( |M\rangle \) with respect to the atomic threshold can be realized by changing the modulation characteristics. For the different cases, the damping and energy shift of the discrete state are evaluated [29]. The results allow the characterization of the time scale where, despite the existence of damping, a coherent response can be assumed. In that scale, the effective discretization, which requires the modification of the coupling constant to incorporate the properties of the density of states and the confining volume [30], is sound. One should take into account that in this approach, the variation of the energy \( E_A \) along the atomic set is still contemplated. This is particularly important to describe experiments done on uncondensed samples [18, 19], where the starting point is a thermal distribution of atoms. The variation of \( E_A \) in the distribution of atomic energies implies dealing with a continuous range of frequencies in the system response, and therefore, with the emergence of dephasing effects when the averaging over the distribution is carried out. The characteristic time for the associated decoherence is determined by the width of the state distribution, and, therefore, by the temperature.

A second line of generalization of our model refers to the inclusion of many-body effects. An operative reduction of the complete microscopic dynamics must incorporate two fundamental elements. First, for a condensate preparation, since, each of the \( N \) identical atoms of the system can interact with \( N - 1 \approx N \) others to form the molecular species, a scaling of the interaction strength in the equations for the evolution of the populations is needed. This bosonic stimulation can significantly affect the efficiency of the population transfer. Second, the correct procedure to renormalize the dynamical equations must take into account that the number of single atoms in the system changes as the conversion progresses. This implies the inclusion of an effective interaction strength which depends on the (evolving) atomic population, and, therefore, the use of nonlinear equations. (The generalization along this line and the inclusion of the dependence of the interaction strength on the confining volume are crucial for understanding the relevance of the atom density to the conversion efficiency.) Following these lines, our former (simplified) description of the system is replaced by a nonlinear Rabi model [1, 2, 30].

Apart from many-body physics, the model must incorporate molecular decay if the time scale for this process is smaller than that of the complete conversion transfer. Losses can be incorporated in a phenomenological way: they can be simply described as a depletion of the molecular population characterized by a constant rate [20, 22]. This implies dealing with a damped nonlinear Rabi model.

Previous reports on atom-molecule conversion methods have stressed the technical difficulties of avoiding magnetic
non-uniformity in the practical setups [27]. Hence, in addition to damping due to dissociation and thermal decoherence, it is worth considering the spatial variations in the applied magnetic field as a possible source of dephasing. Our approach provides a framework where the inhomogeneities can be straightforwardly incorporated: in a local-density approximation, a nonuniform magnetic field can be tackled by simply averaging the results previously obtained for the homogeneous case over the distribution of field values.

Obviously, the above elements are not specific to the proposed setup. They are present in the standard scenario defined by a single-frequency modulation field, where, despite their effects, there is a significant time scale where a coherent response is observed. Then, we can infer that, although those components can affect the system dynamics, they do not preclude the applicability of the interference scheme.

A source of decoherence specific to the interference scheme is that rooted in field-phase fluctuations. The precise fixing of the phase difference between the modulation-field components is essential for the method to work. We can incorporate phase noise into our framework through a distribution centered on the optimum phase value and with a width depending on the particular setup conditions. In our reduced picture, the evolution of the atomic and molecular populations corresponds to a Rabi oscillation, (linear or nonlinear, depending on the magnitude of many-body effects), with the frequency being determined by the effective interaction strength and by the detuning. Then, fluctuations in the phase lead to the random variation of the frequency, and, consequently, to dephasing in the output oscillations. The time scale where coherence persists depends then on the width of the noise distribution. Again, it seems that, in standard arrangements, reduced levels of phase noise can be realized.

5. Concluding remarks

We have shown that the interference between association paths can provide a tool for controlling the conversion of ultracold atoms into molecules in the magnetic-field modulation scenario. The coherence required for the interference scheme to work seems to be feasible in standard arrangements. Actually, although the simplified picture used to present the method of control can be considerably modified when different components of the system, (e.g., many-body effects, molecular decay, decoherence, or nonuniformities in the field) are incorporated, the central effects generated by the interference persist. Namely, the induced modification of the interaction strength is present in any required generalization of the applied approach. The applicability of the study to a system of fermions, in particular, the differential treatment of many-body effects, will be the subject of future work.

The second main goal of our study has been the characterization of second-order effects in the field modulation. We have incorporated into the basic model the quadratic dependence of the binding energy of the FR state on the driving field. We have shown that the combined effects of linear and quadratic terms can be regarded as a particular case of the proposed interference scheme. Actually, those effects have been described using the general approach previously developed to deal with control issues. As a result, we have explained the shift in the resonance frequency induced by the field quadratic terms, observed in previous experimental work.

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