Quantum effects at low-energy atom–molecule interface

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Abstract. The effects of quantum interference in inter-conversion between cold atoms and diatomic molecules are analysed in this study. Within the framework of Fano’s theory, continuum-bound anisotropic dressed state formalism of atom–molecule quantum dynamics is presented. This formalism is applicable in photo- and magneto-associative strong-coupling regimes. The significance of Fano effect in ultracold atom–molecule transitions is discussed. Quantum effects at low-energy atom–molecule interface are important for exploring coherent phenomena in hitherto unexplored parameter regimes.

Keywords. Continuum-bound dressed state; atom–molecule coherence; photoassociation; magnetic Feshbach resonance; optical Feshbach resonance; Fano interference.

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1. Introduction

Ever since the realization of Bose–Einstein condensation \cite{1–3} in dilute gases of alkali atoms 16 years ago, there has been a revolutionary growth in research activities with cold and ultracold atoms. By ‘cold atoms’, we mean a temperature range for atoms from miliKelvin (mK) down to microKelvin (\(\mu\)K) regimes while ‘ultracold atoms’ imply atoms in the sub-\(\mu\)K or nanoKelvin (nK) temperature regime. Tremendous development in the technology of cooling and trapping of atoms \cite{4–6} in the 1980s and early 1990s culminated in the first demonstrations of the Bose–Einstein condensation \cite{1–3}. The 1997, Nobel prize in physics was jointly awarded to Steven Chu, William D Phillips and Claude N Cohen-Tannoudji for their contributions to laser cooling. Carl E Wieman, Eric A Cornell and Wolfgang Ketterle were awarded the 2001 Nobel prize in physics for the first achievement of Bose–Einstein condensate (BEC) in dilute atomic gases. Over the years, several research groups around the world have demonstrated Bose–Einstein condensation in different alkalies and other kinds of atomic gases. Along with the progress of research in cold bosonic atoms, there has been a lot of advancement in cooling and
trapping of fermionic atoms. Fermi degeneracy in a Fermi gas of alkali atoms was first demonstrated by Jin in 1999 [7].

Cold atoms have revitalized not only atomic physics, but also many other areas of research starting from superfluidity, molecular and condensed matter physics to astrochemistry. This has been possible because of the extraordinary properties of cold atoms making them a fertile ground for studying new physics and chemistry. One of the most interesting properties of cold atoms is the tunability of interatomic interaction over a wide range by an external magnetic field Feshbach resonance (MFR) [8–10]. By changing the strength of an external magnetic field near a Feshbach resonance, one can basically alter s-wave scattering length of ultracold atoms from large positive to large negative values or vice versa. Exactly at resonance, the scattering length diverges. Near Feshbach resonance, an atomic gas becomes strongly interacting. Over the years, MFR has been extensively used as a standard tool for studying strongly interacting atomic Fermi gases [11–13]. In fact, it has facilitated to achieve a number of milestones in the area of Fermi gases: (1) realization of s-wave fermionic superfluidity [14] in a Fermi gas of alkali atoms, (2) achievement of molecular Bose–Einstein condensation [15–17] of a new type of diatomic molecules known as ‘Feshbach’ molecules formed from cold fermionic atoms and (3) demonstration [14,18,19] of a cross-over between Bardeen–Cooper–Schrieffer (BCS) state and BEC known as BEC–BCS or BCS–BEC cross-over [20,21]. All these progresses along with the development of laser-generated artificial crystalline structures for cold atoms known as optical lattices have made cold atoms a test bed for the models of condensed matter physics. One of the advantages for simulating fundamental quantum phenomena of condensed matter systems with cold or ultracold atoms is that, unlike electrons in solid-state materials, cold atoms allow unprecedented control over atom–atom interaction. New insight into the quantum physics of many-body systems can be developed to enrich our understanding on unsolved problems in physics such as high-temperature superconductivity [22]. On the one hand, cold atom science offers new opportunities for quantum simulation of condensed matter phenomena; on the other hand, research into quantum optical phenomena with interacting cold or ultracold atoms is opening up new vistas of research into hitherto unexplored quantum effects at the interface of atomic and molecular states. The advent of Feshbach molecules due to MFR and the formation of cold molecules by photoassociation (PA) [23–25] have generated a lot of research interest in physics as well as chemistry. Both the PA and MFR methods have several common features. An analogy of PA with MFR has been exploited to develop an optical method of altering s-wave scattering length known as optical Feshbach resonance (OFR) [26–28]. Although OFR has so far not been as effective as MFR as far as tuning of s-wave scattering length is concerned, optical methods based on strong-coupling PA with quantum interference between optical transition pathways has been theoretically shown to be useful for altering higher partial-wave atom–atom interaction [29].

In this review article, we present an overview on coherent phenomena involving atom–molecule transitions at low energy with particular focus on quantum interference effects at atom–molecule interface (AMI). It is almost impossible to make such a review self-content given the diversity of current state-of-the-art research with ultracold atoms and molecules. Numerous excellent reviews, monograms and books on cooling and trapping of atoms, precision spectroscopy, Bose–Einstein condensates and related subjects are written over the last one and a half decade. It is not our aim to give an overview
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of these topical subjects that are well-documented in the literature. Instead, we refer to some reviews and books [30–33] for general readers. Our discussions are mainly centred around some selective theoretical topics related to interconversion between atoms and molecules at ultralow energy in the presence of external fields. Ultracold molecules are an emerging area of research with tremendous prospects for exploring new physics and chemistry. In recent times, a few focus or special articles have appeared in scientific magazines and journals revealing the broad and general interest of research with ultracold molecules. For instance, we can cite the recent article by Jin and Ye [34], the review by Carr et al [35] and the book on cold molecules [36].

In §2, after a brief discussion on the formation of molecules from cold or ultracold atoms, continuum-bound spectroscopy is described with emphasis on the underlying processes of association of two atoms into a molecule. The processes discussed are PA and MFR. Section 2 provides a general discussion on the associative processes while the technical aspects of PA and MFR are described in §3 and 4. In §3, continuum-bound dressed state formalism is presented for describing quantum dynamics at AMI. Section 4 is primarily devoted to the analysis of quantum interference phenomena that can arise in atom–molecule transitions. In case of multiple pathways of dipole transitions at AMI, quantum interference may arise between two competing pathways. These interference effects can be controlled by an external static or dynamic magnetic or electric field or combination of several fields. In §5, conclusion and a brief discussion on prospects of new physics and chemistry at AMI are given.

2. Continuum-bound spectroscopy: Photoassociation and Feshbach resonance

Photodissociation of diatomic molecules is a well-known bound-continuum process where a molecule is fragmented into its constituent atoms due to absorption of light. Photodissociation spectroscopy has been long studied and developed into a standard method of bound-continuum or continuum-bound spectroscopy. In contrast, PA is a relatively new kind of continuum-bound spectroscopy developed mostly over the last 25 years in the modern era of laser cooling and trapping of atoms much below 1 K. PA is a process by which two colliding ground-state cold atoms in the presence of laser light of appropriate frequency can be associated to form an excited diatomic molecule via absorption of one photon. So, this is a dipole transition from the continuum (two unbound ground state atoms in the state of collision) of ground diatomic molecular potential to the ro-vibrational bound states in the electronically excited molecular potential. Excited molecule formed by PA is long-ranged compared to the typical range of diatomic molecules (≤ 20\(a_0\), where \(a_0\) is the Bohr radius) at room temperature. The binding energies of excited molecules formed by PA lie very close to the dissociation threshold of the molecular potential – typically in the range of sub-mK to several hundred mK below the threshold. Therefore, the initial temperature of the atomic cloud should be cold enough to access such loosely bound molecular states by PA. While dissociation of a diatomic molecule by an external perturbation is possible at room temperature, associating two colliding atoms into a diatomic bound state requires cold temperature because of the nature of molecular continuum-bound spectroscopy that we shall discuss shortly. This is why associating
spectroscopy due to an external field had not developed into a full-fledged discipline until the advent of cold atoms.

Not only PA spectroscopy, but advancement in cold atom research has also led to another new kind of continuum-bound spectroscopy due to MFR. While PA is due to electric dipole continuum-bound transitions involving excited electronic configurations, MFR spectroscopy is based on magnetic coupling between continuum and a quasibound state at near-zero collision energy with quasibound and continuum states belonging to different configurations (scattering channels) of two ground state atoms. MFR spectroscopy has led to the emergence of Feshbach molecules which are produced due to three-body collisions near the magnetic field at which two-body scattering (Feshbach) resonance occurs. These molecules are translationally and rotationally cold. Since their binding energies are close to the threshold of ground molecular potentials, they have extremely high vibrational quantum numbers. If a Feshbach molecule is formed from a pair of two-component fermionic atoms, the molecule attains extraordinary stability, because inelastic molecule–atom or molecule–molecule scattering is suppressed due to Pauli blocking. With magnetically tunable Feshbach resonance method, two-component Fermi gases of ultracold atoms are convertible into Bose–Einstein condensates of Feshbach molecules.

The experimental study of PA in the presence of MFR was initiated in the late 1990s [37–41]. Over the last four years, several such experiments [42–46] have been carried out. The results of these experiments as well as theoretical studies [47–53] reveal significant effects of magnetic field-induced Feshbach resonances on PA and cold collision properties. According to Franck–Condon principle, the intensity of bound–bound (molecule–molecule) or continuum-bound (dissociative or associative) spectrum is in general proportional to the square of overlap integral between the two bound states or between continuum and bound states. Therefore, a continuum-bound transition is most probable when the most prominent (with the largest amplitude) antinodes of continuum and bound states are located at interatomic separations which are nearly equal or comparable. When the antinode of either continuum or bound state lies at a separation close to the node of the other, the probability of transition between such continuum and bound states is the least. In the presence of Feshbach resonance, enhancement and suppression in PA spectral intensity profile can take place due to quantum interference between PA and Feshbach resonances leading to Fano-type asymmetric spectral profile. Such quantum interference at AMI has important implications in coherent control of atom–atom cold collision and bound-state properties. For instance, quantum interference in the strong PA coupling limit leads to nonlinear Fano effect and suppression of power broadening in PA spectrum, double resonances in atom–atom scattering and manipulation of higher partial-wave interaction. Studies on quantum interference at AMI may lead to another new kind of continuum-bound spectroscopy, which we may call as Fano–Feshbach quantum interference spectroscopy (FFQIS).

PA spectrum is usually determined by measuring the loss of atoms from atomic traps due to a PA laser. Once a bound state is formed in an excited molecular potential by PA process, this bound state can spontaneously decay to either two hot atoms or a bound state in ground molecular potential close to the dissociation threshold. These hot atoms or ground molecules in highly excited vibrational levels then escape from the trap. Theoretical treatment of PA spectrum views PA process as laser-assisted or laser-modified atom–atom scattering with spontaneous decay from the excited bound states. Bohn and Julienne
[54,55] have developed a semianalytical theory of PA spectroscopy based on multichannel scattering theory with an artificial lossy channel. The characteristic features of PA spectroscopy as trap loss can be drastically changed mainly in three situations: (1) strong-coupling PA regime, that is, a regime where PA dipole coupling exceeds spontaneous emission linewidth, (2) interference of MFR with PA and (3) application of multiple lasers driving multiple continuum-bound dipole transitions in the strong-coupling regime.

In strong photoassociative dipole coupling [56,57], rate of stimulated bound-continuum transition can take over the rate of spontaneous transition recycling the atoms into the initial continuum thereby stalling trap loss. As a result, atom–molecule coherence between the ground continuum and the excited molecular state can emerge forming a continuum-bound ‘dressed state’. It is then appropriate to describe PA dynamics in the framework of a dressed state picture. Fifty years back, in a classic paper, Fano [58] showed how to diagonalize an interacting continuum-bound system resulting in ‘dressed continuum’. In the late 1950s, Feshbach [59] formulated the mathematical theory of a particular type of two-body scattering resonances now known as ‘Feshbach resonance’ in literature. For Feshbach resonance to occur, three physical conditions need to be satisfied: (i) existence of a quasibound state supported by a closed scattering channel is needed, (ii) a coupling between these closed and open channels is required and (iii) initial collision energy has to be close to the binding energy of the quasibound state. In multichannel two-body scattering theory, a channel is defined by a possible arrangement of internal degrees-of-freedom (such as electronic or hyperfine spin, angular momentum of relative motion, etc.) in the asymptotically large separation limit of two bodies. In essence, both Fano and Feshbach methods deal with a continuum interacting with one (more than one) bound state (states). Therefore, both methods are related, although the formalism and the physical contexts in which both methods were originally developed are quite different. In the absence of any loss or inelastic process, both methods provide exact treatments of an interacting continuum-bound system and therefore they are valid in any coupling regime. PA is usually viewed as a trap loss spectroscopy studied mostly in the weak-coupling regime, although PA theory has made use of Fano’s method [54,55]. Description of strong-coupling PA requires an explicit dressed state picture based on Fano–Feshbach diagonalization method. There are several interesting strong-coupling effects which include (a) large shifts and linewidth exceeding spontaneous linewidth, (b) occurrence of multiphoton process, (c) generation of higher partial-wave interaction in the continuum and (d) excitations of higher rotational levels in the excited molecules.

When PA occurs in the presence of MFR, the existence of a closed channel quasibound state opens up a bound–bound dipole transition pathway due to PA laser. In such a physical situation, there are three competing couplings of which two are continuum-bound types and one is the bound–bound type. Of the two continuum-bound couplings, one is the PA coupling and the other is the magnetic one between the closed channel bound state and the ground scattering state. Naturally, quantum interference arises between any two of these three possible transition pathways. Strong-coupling phenomena are then largely influenced by these quantum interference effects. Theoretical treatment of quantum interference in strong-coupling regime of PA rests on the method of Fano–Feshbach diagonalization resulting in dressed continuum. In what follows, we show that quantum interference at AMI can significantly manipulate molecule formation and continuum...
In §3, we present the basic formalism for obtaining dressed continuum that is central to the description of quantum interference effects at AMI as discussed in §4.

3. Dressed state formalism of continuum-bound quantum dynamics

As mentioned in the preceding section, following Fano or Feshbach method, one can diagonalize an interacting system of continuum and bound states to obtain dressed continuum. The method we follow to diagonalize a PA system is partly a synthesis of the two methods. Our method is based on real-space Green functions that are used to solve second-order coupled differential equations. Since our interest lies in the manipulation of bound and scattering states by strong-coupling effects, we prefer to work in coordinate space to derive explicitly scattering amplitudes that can be determined from asymptotic analysis of dressed continuum at large separations. Let us consider PA process as schematically depicted in figure 1. To begin with, let us idealize PA system by assuming that the excited bound state is loss-less, that is, it has infinite lifetime. Spontaneous decay of the excited state will be introduced later. Let the PA laser be tuned near resonance of a particular ro-vibrational bound state characterized by vibrational quantum number $v$ and rotational quantum number $J$. Generally, $J$ is given by $J = L + S + \ell$, where $L = l_a + l_b$ is the total electronic orbital angular momentum of the two atoms with $l_a$ being the atomic electronic orbital angular momentum of $\alpha (a, b)$-atom, $S$ is the total electronic spin angular momentum and $\ell$ represents the angular momentum of the relative motion of the two atoms. In case of bound–bound spectroscopy of diatomic molecule, angular momentum of relative

![Figure 1](image_url)

**Figure 1.** A schematic diagram of PA, showing ground and excited state adiabatic molecular potentials. Singlet and triplet ground-state potentials are mixed up due to hyperfine interaction. PA laser excites two ground-state ($S$) atoms colliding in the continuum of molecular ground-state potential into a bound state in the excited molecular potential which asymptotically corresponds to one ground ($S$) and another excited ($P$) atom.
motion or rotation of internuclear axis is usually represented by the symbol $N$ instead of $\ell$.

For simplicity, let us assume that the two ground-state atoms collide in a single channel (configuration of internal degrees-of-freedom of the two separated atoms) denoted by $|g\rangle = |f_a, f_b, F\rangle$, where $f_{a(b)}$ is the hyperfine quantum number of atom $a(b)$ and $F = f_a + f_b$. Let the angular state of the excited bound state be denoted by $|e\rangle$, which includes all possible angular quantum numbers to characterize the excited state including the rotational quantum number $J$ and its projection $\zeta$ on the body-fixed $z$-axis, which is the internuclear axis. Under Born–Oppenheimer approximation, integrating over electronic coordinates, the Hamiltonian of the system can be written as

$$H = H_{\text{rel}} + H_{\text{CM}},$$

where $H_{\text{CM}} = -(\hbar^2/2M)\nabla^2_{R_{\text{CM}}}$ describes the centre-of-mass motion of the two atoms with total mass $M = m_a + m_b$, where $m_{a(b)}$ is the mass of the atom $a(b)$, $\nabla_R$ represents Laplacian corresponding to the centre-of-mass position vector $R = (m_a\vec{r}_a + m_b\vec{r}_b)/M$ with $\vec{r}_{a(b)}$ being the position vector of atom $a(b)$. The Hamiltonian of relative motion of the two atoms is given by

$$H_{\text{rel}} = -\frac{\hbar^2}{2\mu} \nabla^2_r + \sum_{\alpha=g,e} (E_{\alpha} + V_{\alpha}) |\alpha\rangle \langle \alpha| + (e^{-i\omega_L t} V_{\text{int}}^{(PA)} |e\rangle \langle g| + \text{c.c.}),$$

(1)

where $\nabla_r$ is the Laplacian corresponding to the relative position vector $\vec{r} = \vec{r}_a - \vec{r}_b$, $\mu = m_a m_b/M$ is the reduced mass and $\omega_L$ is the angular frequency of PA laser. Here, $E_{\alpha}$ is the asymptotic internal (electronic) energy of the molecular state $|\alpha\rangle$, that is, total internal energy of the two atoms at large separation (separated atom limit). $V_{\alpha}$ denotes the atom–atom interaction in state $\alpha = g, e$ and $V_{\text{int}}^{(PA)} = -\vec{D} \cdot \vec{E}$ is the PA laser-induced dipole interaction, where $\vec{D}$ is the molecular transition dipole moment and $\vec{E}$ is the electric field of PA laser. The ground molecular state ($\alpha = g$) at large separation corresponds to two $S$ ($l_a = l_b = 0$) atoms and hence, we can set $E_g = 0$, while the excited state ($\alpha = e$) in the separated atom limit corresponds to one $S$ and another $P$ (either $l_a$ or $l_b$ being equal to 1) and so $E_e = \hbar \omega_B$, where $\omega_B$ is the atomic transition frequency between $S$ and $P$ states. Since $H_{\text{CM}}$ describes only the free motion of the centre-of-mass of the two atoms, the wave function is separable in two parts—centre-of-mass and relative wave functions. The centre-of-mass motion does not influence atom–atom scattering at all and introduces only a phase factor $\exp[i\vec{K} \cdot \vec{R}]$ to the total wave function. Henceforth, we therefore discuss only the relative motion of the two atoms.

The time-dependent relative wave function can be formally expressed as $|\Psi(t)\rangle = e^{-iH_{\text{rel}}t/\hbar} |\Psi(t = 0)\rangle$. Suppose, the Hamiltonian $H_{\text{rel}}$ satisfies the eigenvalue equation $H_{\text{rel}} |E\rangle_{dr} = E |E\rangle_{dr}$, where $|E\rangle_{dr}$ is the eigenfunction known as dressed continuum with $E$ being the eigenenergy. These dressed basis are energy-normalized, that is, $\langle E’|E\rangle_{dr} = \delta(E - E’)$. One then defines an identity operator $I = \int dE |E\rangle_{dr} \langle E|$. Making use of this identity operator, $|\Psi(t)\rangle$ can be expanded in terms of these dressed continuum basis in the following form:

$$|\Psi(t)\rangle = \int dE e^{-iEt/\hbar} |E\rangle_{dr} \langle E|\Psi(t = 0)\rangle.$$

(2)

Let the rotational energy spacing of the excited ro-vibrational states be much larger than PA laser linewidth such that the laser can effectively couple only a single rotational level.
In the absence of any external magnetic field, the excited ro-vibrational level has \((2J + 1)\)-fold degeneracy. We therefore need to consider dressed state of \((2J + 1)\) degenerate bound states interacting with a single continuum. The dressed continuum for such an interacting system can be expressed as

\[
|E\rangle_{dr} = \sum_{\ell M} A_{JM}(E, \hat{k})|b\rangle|e(J\zeta M)\rangle + \sum_{\ell m_{\ell}} \int dE' C_{E',\ell m_{\ell}}(E, \hat{k})|E'\ell m_{\ell}\rangle_{\text{bare}}|g\rangle,
\]

(3)

where \(M\) is the projection of \(J\) on the space-fixed (laboratory frame) \(z\)-axis, \(\hat{k}\) denotes the direction of the incident and scattered relative momenta; \(|b\rangle\) and \(|E\rangle_{\text{bare}}\) are the bare bound (ro-vibrational) and bare continuum states, respectively. \(A_{JM}(E, \hat{k})\) and \(C_{E}(E, \hat{k})\) are the laser-dependent coefficients to be determined. \(A_{JM}(E, \hat{k})\) can be expanded in the form \(A_{JM}(E, \hat{k}) = \sum_{\ell m_{\ell}} A_{JM}^{\ell m_{\ell}}(E, \hat{k})\) and similarly \(C_{E}(E, \hat{k}) = \sum_{\ell m_{\ell}} C_{E\ell m_{\ell}}(E, \hat{k})\).

The coordinate representation of the dressed continuum is \(|r|E\rangle_{dr} = \Psi_{\hat{k}, E}(r)\) and that of bare bound state is \(|r|b\rangle|e(J\zeta M)\rangle = |r|b\rangle|e(J\zeta)\rangle = r^{-1}\phi_{\ell \mu J}(r)|J\zeta M; \eta\rangle\), where

\[
|J\zeta M; \eta\rangle = i^{J} \sqrt{\frac{2J + 1}{8\pi^{2}}} D_{M\zeta}^{(J)}(\hat{r})|\eta\rangle.
\]

(4)

Here, \(D_{M\zeta}^{(J)}(\hat{r})\) is the Wigner rotation matrix element and \(\hat{r}\) represents the Euler angles and \(|\eta\rangle\) denotes the electronic or hyperfine spin angular state of the excited bound level. Because of the cylindrical symmetry of diatomic molecules with respect to the internuclear axis, \(\zeta\) is a good quantum number. Let us assume that the excited molecular state belongs to Hund’s case \(a\) or \(b\). Then \(\Lambda\) which is the projection of \(L\) on internuclear axis is also a good quantum number. For simplicity, we consider that the excited state has \(\Sigma\) symmetry, that is \(\Lambda = 0\) and \(S = 0\) (spin singlet). For \(\zeta = 0\), \(|J\zeta M; \eta\rangle\) reduces to \(Y_{JM}(\hat{r})|\eta\rangle\) with \(Y_{JM}(\hat{r})\) being the spherical harmonics. Similar to the dressed continuum, the bare continuum states are also energy-normalized. In the cold collision regime, the lowest partial wave \(\ell = 0\) (s-wave) is the most significant and a few higher partial waves (such as \(p\)-, \(d\)-wave and so on) may have finite contributions depending on the temperature of the cold atomic cloud. It is advantageous to expand the coordinate representation of bare continuum \(|r|E', \ell m_{\ell}\rangle_{\text{bare}} = \frac{1}{r}\psi_{E, \ell m_{\ell}}(r)Y_{\ell m_{\ell}}(\hat{r})\), where the wave function \(\psi_{E, \ell m_{\ell}}(r)\) satisfies the time-independent Schrödinger equation

\[
\left[ \frac{\hbar^{2}}{2\mu} \left\{ -\frac{d^{2}}{dr^{2}} + \frac{\ell(\ell + 1)}{r^{2}} \right\} + V_{g}(r) \right] \psi_{E, \ell m_{\ell}}(r) = E \psi_{E, \ell m_{\ell}}(r).
\]

(5)

If the ground potential \(V_{g}(r)\) is spherically symmetric, then \(\psi_{E, \ell m_{\ell}}(r)\) becomes independent of \(m_{\ell}\). Note that, although bare continuum can be isotropic in case of spherically symmetric ground-state atom–atom potentials, dressed continuum becomes anisotropic due to PA coupling which is essentially anisotropic. In fact, any external field-modified scattering is anisotropic in general. Therefore, we need to incorporate the mathematical formulation of anisotropic scattering within the framework of Fano’s theory for a description of strong-coupling PA. Using the coordinate representations of bare excited bound
and ground continuum states, we can express the dressed continuum of eq. (3) in the following form:

\[
\Psi_E(\hat{k}, \mathbf{r}) = \frac{1}{r} \left[ \sum_M \Phi_{E,vJ}(\hat{k}, r)|J\xi M; \eta) + \int dE' \sum_{\ell m_{\ell}} \Psi_{E',\ell m_{\ell}}(\hat{k}, E, r)|\ell 0m_{\ell}; g) \right],
\]

where

\[
\Phi_{E,vJ}(\hat{k}, r) = \sum_{\ell m_{\ell}} A_{JM}^{\ell m_{\ell}}(E)\phi_{vJ}(r) Y_{\ell m_{\ell}}(\hat{k}),
\]

\[
\Psi_{E',\ell m_{\ell}}(E, \hat{k}, r) = \sum_{\ell' m_{\ell'}} C_{E'\ell m'\ell m_{\ell}}(E)\psi_{E',\ell m_{\ell}}(r) Y_{\ell' m'\ell m_{\ell}}(\hat{k})
\]

(7)

and \(|\ell 0m_{\ell}; g) = |\ell 0m_{\ell}; g)\). In what follows, we calculate the coefficients \(A_{JM}^{\ell m_{\ell}}\) and \(C_{E'\ell m'\ell m_{\ell}}(E)\) by solving the two coupled differential equations by the method of Green function.

Substituting eq. (6) in time-independent Schrödinger equation \(H_{\text{rel}}\psi_E(\mathbf{r}) = E\psi_E(\mathbf{r})\), we obtain the following two coupled differential equations:

\[
\left[ -\frac{\hbar^2}{2\mu} \frac{d^2}{dr^2} + B_J(r) + V_e(r) - \hbar \delta_L - E \right] \Phi_{E,vJ} = -\sum_{\ell m_{\ell}} V^{PA}_{JM,\ell m_{\ell}}(r) \tilde{\psi}_{E,\ell m_{\ell}}
\]

\[
\left[ -\frac{\hbar^2}{2\mu} \frac{d^2}{dr^2} + B_\ell(r) + V_g(r) - E \right] \tilde{\psi}_{E,\ell m_{\ell}} = -\sum_M V^{PA}_{\ell m_{\ell};JM}(r) \Phi_{E,vJ},
\]

(8)

(9)

where \(\delta_L = \omega_L - \omega_A\) is the laser–atom detuning, \(B_J(r) = \hbar^2/(2\mu r^2)J(J + 1)\) is the rotational term of the excited molecular bound state, \(B_\ell(r) = \hbar^2/(2\mu r^2)\ell(\ell + 1)\) is the centrifugal term in collision of two ground-state atoms and \(V^{PA}_{JM,\ell m_{\ell}} = \langle \ell 0m_{\ell}; g)|V^{|J\xi M; \eta)\rangle\) is the electronic coupling matrix element for the transition \(|J\xi M; \eta)\rightarrow|\ell 0m_{\ell}; g\rangle\). Here \(\tilde{\psi}_{E,\ell m_{\ell}} = \int dE'\psi_{E',\ell m_{\ell}}(E, r) = \int dE'\sum_{\ell' m'_{\ell}} C_{E'\ell m'\ell m_{\ell}}(E)\psi_{E',\ell m_{\ell}}(r)\). The coupling \(V^{PA}_{JM;\ell m_{\ell}}\) depends weakly on internuclear separation \(r\).

The two coupled eqs (8) and (9) reduce to two separated homogeneous equations in the absence of PA laser \((V^{PA}_{\text{int}} = 0)\). Therefore, the coupled equations are exactly solvable by the techniques of Green function provided the solutions of the corresponding homogeneous equations are known. Let the bound state solution of the homogeneous part of eq. (8) with energy \(E_{vJ}\) be represented by \(\phi_{vJ}(r)\). The corresponding Green function is

\[
G_v(r, r') = -\frac{\phi_{vJ}(r')\phi_{vJ}(r)}{\hbar \delta_L + E - E_{vJ}}.
\]

(10)

The Green function for the homogeneous part of eq. (9) can be expressed as

\[
\mathcal{K}_{E,\ell}(r, r') = -\pi \left[ \psi_{E\ell}^{\text{reg}}(r)\psi_{E\ell}^{\text{irr}}(r') + i \psi_{E\ell}^{\text{reg}}(r)\psi_{E\ell}^{\text{reg}}(r') \right], \quad r' > r
\]

\[
\mathcal{K}_{E,\ell}(r, r') = -\pi \left[ \psi_{E\ell}^{\text{reg}}(r')\psi_{E\ell}^{\text{irr}}(r) + i \psi_{E\ell}^{\text{reg}}(r')\psi_{E\ell}^{\text{reg}}(r') \right], \quad r' < r,
\]

(11)

(12)
where $\Psi_E^{\text{reg}}(r)$ and $\Psi_E^{\text{irr}}(r)$ represent the regular and irregular scattering solutions in the absence of laser field. $\Psi_E^{\text{reg}}(r)$ goes to zero at $r = 0$, while $\Psi_E^{\text{irr}}(r)$ is defined by boundary condition at $r \to \infty$ only. Following the procedure as described in ref. [56], we can expand $A_{JM}(E) = \sum_{\ell m_\epsilon} A_{JM, \ell m_\epsilon}$ with

$$A_{JM, \ell m_\epsilon}(E) = \frac{\Lambda_{JM, \ell m_\epsilon}(E)}{\hbar \delta_L + E - (E_{vJ} + E_{vJ}^{\text{shift}}) + i\hbar \Gamma_{JM}(E)/2},$$

where

$$\Lambda_{JM, \ell m_\epsilon}(E) = \int \phi_{vJ}(r)V_{JM, \ell m_\epsilon}(r)\Psi_{E, \ell m_\epsilon}(r)dr$$

is the continuum-bound molecular dipole matrix element. Here $\Gamma_{JM} = \sum_{\ell m_\epsilon} \Gamma_{JM, \ell m_\epsilon}$ with

$$\Gamma_{JM, \ell m_\epsilon}(E) = \frac{2\pi}{\hbar} |\Lambda_{JM, \ell m_\epsilon}(E)|^2$$

being the bound-continuum partial stimulated linewidth. The total shift $E_{JM}^{\text{shift}} = \sum_{\ell m_\epsilon} E_{JM, \ell m_\epsilon}^{\text{shift}}$ where the partial shift is given by

$$E_{JM, \ell m_\epsilon}^{\text{shift}} = \int \int dr'dr \phi_{vJ}(r')V_{JM, \ell m_\epsilon}(r')\text{Re}[\mathcal{K}_E(r', r)]V_{JM, \ell m_\epsilon}(r)\phi_{vJ}(r).$$

The coefficient $A_{JM, \ell m_\epsilon}(E)$ can be expressed in a compact form as

$$A_{JM, \ell m_\epsilon}(E) = \left[ \frac{\Lambda_{JM, \ell m_\epsilon}(E)}{\hbar \Gamma_{JM}(E)/2} \right] \frac{1}{\epsilon_L + i},$$

where $\epsilon_L = [E + \hbar \delta_L - (E_{vJ} + E_{vJ}^{\text{shift}})/(\hbar \Gamma_{JM}(E)/2)$. Thus, with explicit analytical form of $A_{JM, \ell m_\epsilon}(E)$, we have $\Phi_{E, vJ}(r) = \sum_{\ell m_\epsilon} A_{JM, \ell m_\epsilon}(E)\phi_{vJ}(r)$. Substituting this in eq. (9), and making use of Green function of eqs (11) and (12), we obtain

$$\tilde{\Psi}_E^{\ell m_\epsilon}(E, r) = \psi_E^{\text{reg}}(r) - \sum_{\ell m_\epsilon'} \int dE' \sum_M \tilde{A}_{JM, \ell m_\epsilon}'(E)
\times \int dr' \mathcal{K}_E^{\ell m_\epsilon}(r, r')V_{JM, \ell m_\epsilon}(r')\phi_{vJ}(r').$$

Now, comparing the above expression with $\tilde{\Psi}_E^{\ell m_\epsilon}(E, r) = \sum_{\ell m_\epsilon'} \int dE' C_{E', \ell m_\epsilon}^{\ell m_\epsilon'}(E)\psi_E^{\ell m_\epsilon}(r)$ we get

$$C_{E', \ell m_\epsilon}^{\ell m_\epsilon'}(E)\psi_E^{\ell m_\epsilon}(r) = \delta(E - E')\delta_{\ell, \ell'}\delta_{m_\epsilon, m_\epsilon'} \psi_E^{\text{reg}}(r)
- \sum_M A_{JM, \ell m_\epsilon}(E) \int dr' \mathcal{K}_E^{\ell m_\epsilon}(r, r')V_{JM, \ell m_\epsilon}(r')\phi_{vJ}(r').$$

Equation (18) is an explicit analytical expression for anisotropic dressed scattering state in partial-wave basis in an ideal loss-less situation. The effect of spontaneous emission may
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be incorporated phenomenologically by adding spontaneous linewidth $\gamma_{sp}$ to the stimulated linewidth making the total linewidth $\Gamma_{JM}^{total} = \Gamma_{JM} + \gamma_{sp}$. However, spontaneous emission or any other loss process in continuum-bound system can be introduced from first principle through master equation approach [60,61].

The analytical expressions for scattering $T$- and other matrices can be derived analytically from asymptotic scattering boundary conditions of eq. (18) as described in refs [53,56]. The first term $(\psi_{reg}^{\ell m})$ of partial-wave $(\ell, m)$ scattering state of eq. (18) is the ground-state scattering wave function which is unperturbed by PA coupling, while the effect of PA coupling is embodied in the second term through the amplitude $A_{JM}^{\ell' m'}$, where $\ell'$ and $m'$ correspond to the incident or initial partial wave of unperturbed state. It is possible that one incident partial wave would be scattered into another one due to anisotropic nature of laser coupling. The second term describes the effect of two-photon process – one photon exciting the ground continuum into the bound state and another photon de-exciting it back to the same continuum. Since it is a two-photon process, such effects are possible only in the strong PA coupling regime. As mentioned earlier, at ultracold temperatures only a few low-lying partial waves such as $s$- and $p$-waves are predominantly prevalent in the unperturbed continuum. Accordingly, the selection rules of electric dipole transitions allow PA laser to populate or excite only the lowest or low-lying rotational level or levels in the excited bound state. For instance, $J = 1$ of the bound state can be excited by PA from $s$-wave scattering state. On being de-excited back into the scattering continuum by stimulated transition by the same PA laser, there is a finite probability of $p$-wave ($\ell = 1$) being generated in the continuum [56]. Here, we have restricted our discussion of formalism to one continuum interacting with only one bound state. However, this formalism can be generalized for other physical situations where two bound states interact with a continuum as shown in refs [51–53]. In such situations where multiple transition pathways interfere quantum mechanically, a host of interesting quantum effects are expected to arise. For instance, in V-type bound-continuum–bound atom–molecule interfacial system, vacuum-induced coherence [62] is shown to arise naturally [63]. Furthermore, it is possible to manipulate $d$-wave ($\ell = 2$) interaction efficiently by two lasers driving two PA transitions to two ro-vibrational states [29].

4. Quantum interference effects at atom–molecule interface

Quantum interferences are ubiquitous in physics, especially in quantum optics. Quantum interferences between different atomic transitions have been extensively studied over the years. A host of quantum optical phenomena such as electromagnetically-induced transparency [64], slow light [65,66], vacuum-induced coherence in spontaneous emission [62], etc. arise due to quantum interference between transition pathways. So far, spectroscopic quantum interference phenomena are studied mostly with atoms at room temperature or at a temperature higher than that of ultracold regime. In recent times, quantum interference effect has been demonstrated in molecule formation [67] from cold atoms and Autler–Townes splitting [68,69] in two-photon PA. Another prominent example of quantum interference is that of Fano [58]. The hallmark of Fano interference is the asymmetric line shape originally observed in atomic autoionization spectrum. Eberly [70] described Fano effect as a ‘confluence’ of coherences. For the last 50 years, Fano
effect has been observed in various physical situations [71], although it has been mostly studied in the context of atomic auto- and photoionization spectroscopy, where the continuum belongs to an excited electronic configuration. In Fano’s theory, no spontaneous decay was considered. Agarwal [60,61] introduced spontaneous decay in Fano model through master equation approach and solved the extended Fano model exactly. With the recent advancement in PA, Fano interference with diatomic ground-state continuum has now become important [72]. The processes that underlie Fano interference in PA with MFR and that in an autoionization system are schematically depicted in figure 2. The asymmetric spectral line shape observed by Junker et al [42] may be an indication of the occurrence of Fano-type quantum interference in PA due to a tunable MFR [52].

In the previous section, we have presented a method of deriving real-space anisotropic dressed continuum of a system of one excited bound state interacting via electric dipole interaction with the ground continuum. The same method can be applied to obtain dressed continuum of magnetically-induced Feshbach resonance. Let us consider the simplest two-channel model of MFR. The asymptotic energy (threshold) of the upper (closed) channel is greater than the collision energy $E'$ of the two atoms while the threshold of the lower (open) channel is below the collision energy. The thresholds of the two channels are tunable by an external magnetic field due to Zeeman effect. The closed channel may support a bound state which may lie very close to the open channel threshold. By varying the magnetic field strength, this bound state can be made to move upward or downward just above or near the open channel threshold. The coupling between this bound state and the open channel scattering state occurs due to hyperfine interaction at an intermediate separation (typically between 20 and 30 Bohr radius).

MFR occurs when the collision energy coincides or nearly coincides with the binding energy of the closed channel quasibound state. It follows from the theory of two-body quantum scattering (two-body quantum scattering is essentially the potential scattering of matter-waves) that when a quasibound state is formed by scattering, scattering phase shift as a function of energy varies rapidly through $\pi/2$, implying the occurrence of

![Figure 2. A schematic diagram comparing the mechanism of Fano interference in MFR-modified PA (MFR-PA) system with that of autoionization system (see the text). Note that a single laser can drive the continuum-bound and bound–bound transitions meaning that a single photon can be absorbed or emitted through two competing transition pathways.](image)
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resonance in scattering amplitude or cross-section. Exactly at resonance, the phase shift is $\pi/2$. Since in the Wigner threshold law regime of ultralow energies, atom–atom scattering is dominated by $s$-wave only, the total scattering cross-section in that regime can be approximated to be the $s$-wave cross-section. Wigner threshold laws state that, if the two-body interaction potential is centrally symmetric and short-ranged, implying that the potential has a finite range or falls off exponentially at large separation, then in the limit of energy or wave number ($k$) going to zero, the partial-wave scattering phase shift $\delta_\ell(k)$ behaves as $\delta_\ell(k) \sim k^{2\ell+1}$. These laws further state that, in the case of centrally symmetric long-ranged potentials with inverse power-law ($\sim 1/r^n$, where $n > 2$) behaviour at asymptotically large separation, $\delta_\ell(k) \sim k^{2\ell+1}$ for $\ell \leq (n-3)/2$ and $\delta_\ell(k) \sim k^{n-2}$ for all $\ell > (n-3)/2$. For two spherically symmetric ground state $S$ atoms, the ground-state potential $V_g(r)$ goes as $1/r^6$ (van der Waals type), we have $n = 6$. Let us now see how $s$-wave scattering $a_s$ behaves in the Wigner threshold law regime. $a_s$ may be defined from Bethe’s expansion formula as

$$\lim_{k \to 0} k \cot \delta(k) = -\frac{1}{a_s} + \frac{1}{2} r_e k^2 + \cdots ,$$

(20)

where $r_e$ is an effective range of the two-body potential. The scattering amplitude $f(k)$ is related to $\delta(k)$ by

$$f(k) = -\frac{1}{k} \left[ \frac{1}{\cot \delta(k) + i} \right] ,$$

(21)

which in the limit $k \to 0$ reduces to

$$f(k) \simeq -\frac{a_s}{1 + ika_s} .$$

(22)

Thus for two atoms interacting via the potential $V_g(r)$, in the limit $k$ going to zero and $|ka_s| \ll 1$, $f(k) \simeq -a_s$ becomes a constant. Since two-body effective interaction can be described in terms of scattering amplitude, at ultracold temperatures, the effective two-body interaction of cold atoms can be parametric with $a_s$, provided $a_s$ does not diverge. But this is not the case when resonance occurs. At resonance, $a_s$ diverges and for $|ka_s| \gg 1$, we have $f(k) \simeq i/k$. Near resonance, scattering cross-section has a Lorenzian shape with the inverse of the Lorenzian width being related to the lifetime of the two-body resonance complex, which we call AMI in case of PA. Thus, both PA and MFR have some common features. The differences between the two are: (i) PA involves electronically excited state, but MFR solely occurs in electronic ground configuration, (ii) PA occurs due to laser interaction and so the nature of the continuum-bound coupling is that of electric dipole transition, MFR involves magnetic transitions only, (iii) while MFR can basically tune $s$-wave interaction, OFR is in principle capable of manipulating not only $s$-wave but also higher partial-wave interactions. Based on PA, OFR has been developed for tuning $s$-wave scattering length. Unlike MFR, OFR is found to be an inefficient method as far as tunability of $s$-wave scattering length is concerned. The inefficiency of OFR mainly stems from the inelastic loss due to the spontaneous emission from the excited bound states. Considering the fact that OFR is applicable to all kinds of atoms – whether they are magnetic or non-magnetic, while MFR is applicable only for those atoms which have magnetic moment, it is of interest to devise new methods of suppression of
inelastic loss in PA in order to make PA-based OFR efficient enough for all practical purposes. In attaining that goal, quantum interference at AMI can play an important role as we discuss in what follows next.

For simplicity, let us consider that only s-wave part of the ground continuum is coupled in the excited ro-vibrational state with \( J = 1 \) by PA in the presence of MFR at ultracold temperature. The dressed state of this system can be written as

\[
|E\rangle_{dr} = \sum_{\ell} A_{JM}^{00} |vJM\rangle + B_E |\chi\rangle
\]

\[
+ \int \! dE' C_{E'E\ell=0m\ell=0}(E) |E'\ell = 0 m\ell = 0\rangle,
\]

where \(|\chi\rangle\) represents closed channel quasibound state with \( B_E \) being its amplitude coefficient. The coefficient \( A_{JM}^{00}(E) \) is given by

\[
A_{JM}^{00} = \frac{(q_f + i)/(\epsilon_B + i)}{\tilde{\delta}_L + \epsilon_B \Gamma_f/2 - (q_f - i)^2/(\epsilon_B + i) + i(1 + \tilde{\gamma})/2} \Lambda_{JM}^{00}, \tag{23}
\]

where \( \tilde{\delta}_L = (\delta_L - E_{vJ}/\hbar)/(\Gamma_{0J}^{00}) \), \( \tilde{\Gamma}_f = \Gamma_f/\Gamma_{0J}^{00} \) and \( \tilde{\gamma} = \gamma_{sp}/\Gamma_{0J}^{00} \) with \( \Gamma_{0J}^{00} = 2|\Lambda_{JM}^{00}|^2 \) and \( \gamma_{sp} \) being the stimulated and spontaneous linewidth, respectively, of the ro-vibrational state. Here, \( \Gamma_f(E) = 2\pi \int \! dr \psi_{E,00}^{reg}(r)V_\chi(r)\phi_\chi(r)'|^2 = 2\pi |V_{\chi}|^2 \) is the MFR linewidth, where \( V_\chi(r) \) is the interaction potential between the closed and open channels and \( \phi_\chi(r) \) is the wave function of the closed channel bound state (\( \chi \) state).

\[
\epsilon_B = \frac{E - E_\chi - E_{\chi}^{\text{shift}}}{\Gamma_f/2} \simeq \tilde{E} - B - B_0 / \Delta k a_{bg}, \tag{24}
\]

where \( E_\chi \) is the binding energy of the \( \chi \) state with

\[
E_\chi^{\text{shift}} = \int \! dr V_\chi(r)\phi_\chi(r)' \int \! dr' \text{Re} \langle K_{E,00}(r, r') V_\chi^*(r')\phi_\chi(r') \rangle \tag{25}
\]

being its shift and \( \tilde{E} = E/(\hbar \Gamma_f/2) \) is the applied magnetic field, \( B_0 \) is the magnetic field at which Feshbach resonance occurs at zero energy in the absence of PA laser, \( \Delta \) is the width (in unit of magnetic field) of zero crossing in MFR and \( \Delta k a_{bg} \) is the background scattering length. \( \Delta \) is related to Feshbach resonance linewidth by \( \Gamma_f/2 \simeq \Delta k a_{bg} \delta \mu \Delta \), where \( \delta \mu \) is the difference between the magnetic moment of the \( \chi \) state and the two free atoms in the open channel. \( q_f \) is the well-known Fano \( q \) parameter which in this context is known as ‘Feshbach asymmetry’ parameter. Explicitly,

\[
q_f = \frac{V_{\text{eff}} + \Omega}{\pi \Lambda_{JM}^{00}(E) V_E}, \tag{26}
\]

where \( \Omega \) is the bound–bound Rabi coupling and

\[
V_{\text{eff}} = \text{Re} \int \! dr \phi_{vJ}(r)V_{JM}^{00}(r) \int \! dr' \langle K_{E,00}(r, r') V_\chi^*(r')\phi_\chi(r') \rangle
\]

is an effective interaction between the two bound states as a result of their interactions with the s-wave part of the continuum. \( V_{JM}^{0n}(r) \), \( \Lambda_{JM}^{0n}(E) \) and the Green function \( K_{E,0n}(r, r') \) are defined in §3.
Let us now analyse how eq. (23) can account for Fano profile in MFR-modified PA spectrum. Usually, PA spectrum is obtained by measuring the rate of loss of atoms from the trap due to spontaneous emission from ro-vibrational states. The rate of trap loss at a collision energy $E$ is given by

$$K_{\text{loss}}(E) = \frac{\gamma_{\text{sp}}}{2} \pi \sum_M \sum_{\ell} A_{\ell}^{M}(E)^2,$$

which at ultracold energy can be approximated by neglecting the contributions from all $\ell > 0$. PA spectrum is then given by

$$K_{\text{PA}} = \frac{\gamma_{\text{sp}}}{2} \langle \pi | A_{00}^{00} | v \rangle,$$

where $\langle \cdots \rangle$ implies thermal averaging over distribution of relative velocity $v = \frac{\hbar k}{\mu}$ which is related to the collision energy by $E = \frac{\hbar^2 k^2}{2\mu}$. Fano minimum in the spectrum is given by the condition $\epsilon_B + q_f = 0$ since $A_{00}^{00}$. Assuming $\tilde{E} \ll 1$ (broad Feshbach resonance), we then have $B_{\text{min}} \simeq B_0 - q_f \Delta k a_{\text{bg}}$, where $B_{\text{min}}$ is the value of $B$ at which Fano minimum occurs. If the ro-vibrational state is long-ranged with its outer turning point lying away from that of the $\chi$ state, then from Franck–Condon (FC) principle, it follows that we can neglect the bound–bound Rabi coupling $\Omega_1$. Note that $q_f$ is independent of laser intensity. However, $q_f$ strongly depends on collision energy. Assuming $V_{00}^{00}(r)$ to be $r$-independent, $A_{00}^{00}(E)$ becomes proportional to the FC factor $\eta_{v,J} = \int dr \phi_{v,J}^r(\psi_{E,00}^r)$. At large separation, we have

$$\psi_{E,00}^r(\xi) \simeq \sqrt{\frac{2\mu}{\pi k}} \sin[kr + \delta_{00}^b(k)]$$

In the limit $k \to 0$, $\delta_{00}^b$ may behave as $\delta_{00}^b \simeq -ka_{\text{bg}}$. In case of a broad Feshbach resonance, the background phase-shift $\delta_{00}^b$ may be finite even in the $\mu$K energy regime. This means that for a broad Feshbach resonance, $s$-wave regular scattering wave function in the $\mu$K energy regime cannot have a node at long-range separation. In the limit $k \to 0$, $V_{\text{eff}}$ becomes energy-independent, while $\eta_{v,J} \propto \sqrt{k}$ and $V_E \propto \sqrt{k}$. Therefore, in the limit $k \to \infty$, Fano minimum does not depend on $k$. The Fano minimum $B_{\text{min}}$ crucially depends on $V_{\text{eff}}$ when $\Omega \simeq 0$. The maximum in Fano profile of PA spectrum as a function of magnetic field depends on a pole of $A_{00}^{00}$. There are two poles of $A_{00}^{00}$, one of which may lie close to the Fano minimum and the other one away from the minimum. The second pole is responsible for the maximum.

5. Conclusions and outlook

In conclusion, we have combined Fano’s method with anisotropic scattering theory to develop an ‘anisotropic dressed continuum’ approach for describing the effects of quantum interference between atomic and molecular transitions at low energy. Intensive research is being conducted to form ultracold ground-state molecules in quantum degeneracy limit, in particular polar molecules for their long-range dipolar interactions. Coexistence of atomic and molecular quantum gases will provide an exotic new system for studying many-body quantum physics. Quantum effects at the interface of atomic and molecular states are fundamentally important for quantum control of chemical reactions at low temperatures, and therefore in the long run, ultracold atom–molecule interface in the quantum regime will relate energy research at a basic level.
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