Pumping vortex into a Bose–Einstein condensate of heteronuclear molecules

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Abstract. Heteronuclear molecules have attracted wide attention due to their permanent electric dipole moments. Analogous to atoms with magnetic dipoles, the existence of nonzero electric dipoles significantly enhances the possibilities and mechanisms for the control and design of quantum degenerate molecule systems with electric (E) fields. This work proposes a vortex creation mechanism inside a condensate of heteronuclear molecules through the adiabatic flipping of the axial bias of an analogous E-field Ioffe–Pritchard trap (IPT), extending the original protocol of Isoshima\textit{ et al} (2000 Phys. Rev. A \textbf{61} 063610) for an atomic spinor condensate inside a magnetic (B)-field IPT. We provide both analytic proof and numerical simulations to illustrate the high fidelity operation of this vortex pump protocol. We hope our work provides stimulating experimental possibilities for active investigations in quantum degenerate molecule systems.

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1. Introduction

The impressive progress in the field of atomic quantum gases can be largely attributed to technical advances in the control of atomic quantum states utilizing electromagnetic interactions. While laser cooling with near-resonant light narrows down the atomic momentum distribution, forced evaporation on trapped atoms favors spatial confinement to the quantum degenerate regime. Since the first successful experiment on atomic Bose–Einstein condensation (BEC), or the controlled production of many bosonic atoms into the same quantum ground state of a trapping potential, the field of atomic quantum gases has blossomed into one of the leading frontiers in physics, attracting ever increasing interest. The ultimate goal along this line of thought is to control the state of an arbitrary assembly of interacting many-body systems, and use it either to simulate other physical systems or to perform quantum computations.

Significant efforts have been directed toward the above ambitious goal, with the immediate aim of the more recent endeavors targeted at the generation, control and study of cold molecules. These endeavors include important achievements such as the Bardeen–Cooper–Schrieffer (BCS) to BEC cross-over due to tunable interactions induced through B-field magnetic Feshbach resonances and the production of quantum degenerate heteronuclear molecules in specific low lying ro-vibrational states of their electronic ground states [1]. The latter advance is especially illuminating, highlighting the prospects for observing and utilizing anisotropic and long-range interactions from permanent electric dipoles.

Molecular quantum fluids bring in new possibilities due to their richer internal degrees of freedom, such as alignment and rotation [2], even after their hyperfine spins like those of atomic spinor quantum gases are neglected. In this short paper, we propose a simple mechanism to generate a vortex state, relying on the internal degrees of freedom of a heteronuclear molecule, through adiabatically flipping the axial bias of an external E-field Ioffe–Pritchard trap (IPT) [3]. The physics of our proposal can be analogously mapped onto the successful protocol first proposed [4] and demonstrated for magnetically trapped atomic condensates [5]–[7]. Our theory will hopefully lead to parallel experiments that will begin to address new opportunities afforded by the molecular internal degrees of freedom.

This paper is organized as follows. First, we describe our model for dc E-field trapping of cold heteronuclear molecules based on the interaction between their permanent electric dipoles and a spatially varying static E-field IPT. This is analogous to the operation of the famous IPT for magnetic dipoles with an inhomogeneous B-field [3]. The heteronuclear diatomic molecule is modeled as a three-dimensional (3D) rigid rotor [2]. We then discuss the eigenstructure of a heteronuclear molecule inside a dc E-field and identify states capable of being trapped by a local E-field minimum and their associated quantum numbers. Next, the rotational properties of
such eigenstates are studied, which helps us to illustrate the proposed vortex pump mechanism based on the flipping of the axial bias E-field [4]–[9]. Finally, the conclusion is given.

It is imperative to present this study because this analogy between the E-field and B-field and between molecular E-dipole and atomic B-dipole is not a simple one to map. The magnetic dipole of an atom contains both an electronic and a nuclear component, respectively, proportional to the electronic spin ($\vec{S}$) and the nuclear spin ($\vec{I}$). Its projection onto the local B-field is approximately proportional to the projection of its hyperfine spin $\vec{F} = \vec{S} + \vec{I}$ when the B-field is weak. In contrast, the molecular dipole is a fixed constant and pointed along the molecule axis connecting the two nuclei. The internal rotational angular momentum of the molecule stays in the perpendicular plane of the molecule axis in the absence of an E-field. Despite these differences, our study finds that the Isoshima et al protocol [4]–[6], originally developed for B-field trapped atoms or molecules, remains effective for heteronuclear molecules in an E-field IPT. Many of these comparative studies and analogies will become clear from the materials presented in sections 3 and 4.

2. Trapping a heteronuclear molecule with a static electric field

We consider a diatomic molecule composed of two different atomic nuclei and described in terms of their center of mass $\vec{R}$ and relative coordinate $\vec{r}$. Neglecting the nuclear and electronic spins and their associated interactions, or assuming that we consider an eigenstate of the above internal degrees of freedom, the remaining Hamiltonian reads

$$\mathcal{H} = \frac{\mathbf{P}_R^2}{2M} + \frac{\mathbf{P}_r^2}{2\mu} - \vec{D}(\vec{r}) \cdot \vec{E}(\vec{R})$$

$$= \frac{\mathbf{P}_R^2}{2M} - \frac{\hbar^2}{2\mu r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) + \frac{\mathbf{J}^2(\theta_r, \phi_r)}{2\mu r^2} - \vec{D}(\vec{r}) \cdot \vec{E}(\vec{R}),$$

(1)

where $\vec{E}(\vec{R})$ denotes the inhomogeneous static E-field, which is slowly varying over the molecule size of $r$, and $\vec{D}(\vec{r})$ is the permanent electric dipole moment operator for this specific electronic and internal spin eigenstate [2, 10, 11]. $M = M_1 + M_2$ and $\mu = M_1 M_2/(M_1 + M_2)$ are, respectively, the total and the reduced mass of the two atoms making up the molecule.

We adopt the Bohn–Oppenheimer approximation to study the effective trapping of the molecule’s center of mass motion. Within this approximation, the molecular wave function for $\vec{R}$ and $\vec{r}$ is decomposed into the form $\Psi(\vec{r}, \vec{R}) = \Phi(\vec{r})\psi(\vec{R})$, and we find

$$\left[-\frac{\hbar^2}{2\mu r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial}{\partial r} \right) + \frac{\mathbf{J}^2(\theta_r, \phi_r)}{2\mu r^2} - \vec{D}(\vec{r}) \cdot \vec{E}(\vec{R}) \right] \psi_{M_J}(\vec{r}) = \mathcal{V}_{M_J}(\vec{R}) \psi_{M_J}(\vec{r}),$$

(2)

$$\left(\frac{\mathbf{P}_R^2}{2M} + \mathcal{V}_{M_J}(\vec{R})\right) \Phi_n(\vec{R}) = E^{(n)}_{M_J} \Phi_n(\vec{R}).$$

(3)

Now consider the case of a specific vibrational state, equation (2) above then reduces to the simple form of a 3D rotator in a dc E-field with a rotational constant $B = (\hbar^2/2\mu r^2)$. If we further assume the local E-field direction to be the quantization $z$-axis, then the Schrödinger equation for the internal part of the diatomic molecule becomes

$$\left[BJ^2 - DE(\vec{R}) \cos \theta_r \right] \psi_{M_J}(\theta_r, \phi_r) = \mathcal{V}_{M_J}(\vec{R}) \psi_{M_J}(\theta_r, \phi_r),$$

(4)
where we have neglected the dependence of the permanent dipole moment on the internuclear distance $\vec{r}$.

Some insights can be gained into weak E-fields if a simple perturbation theory is adopted as is done in [11]. We take the unperturbed Hamiltonian to be $H_0 = B J^2$ with $H_0|J, M_J\rangle = E_{JM_J}|J, M_J\rangle$, where the eigenenergy and eigenfunction are, respectively, $E_{JM_J} = BJ(J + 1)$ and $|J, M_J\rangle$, with spherical harmonics $|J, M_J\rangle$ being functions of $\vec{r}$. The dipole interaction $-DE(\vec{R}) \cos \theta$, is treated as a perturbation. To first order the wave function becomes

$$
\psi_{M_J} = |J, M_J\rangle + \frac{DE}{B} \frac{1}{2(J + 1)} \sqrt{(J + 1)^2 - M_J^2} (J + 1, M_J)$$

$$- \frac{DE}{B} \frac{1}{2J} \sqrt{\frac{J^2 - M_J^2}{(2J - 1)(2J + 1)}} |J - 1, M_J\rangle,
$$

and the corresponding eigenenergy to second order in the external E-field becomes

$$
\nu_{M_J} = BJ(J + 1) + \frac{D^2 E^2}{B} \frac{1 - 3M_J^2 / J(J + 1)}{2(2J - 1)(2J + 3)}.
$$

For this model system, $M_J$ is a conserved quantity. Assuming that the center of mass motion is adiabatic with respect to the internal state, the above eigenenergy from the internal state $\nu_{M_J}$ then clearly acts as an effective potential due to its dependence on $\vec{R}$. In particular, we find that any state with $1 - 3M_J^2 / J(J + 1) > 0$ is a weak field seeking state, i.e. free space E-field traps analogous to the B-field IPT can be constructed. These states, of course, are metastable, and trapping is possible because of the dynamic stability from the Larmor-like precession of the permanent electric dipole along the direction of a local E-field. On the other hand, the strong field seeking states with $1 - 3M_J^2 / J(J + 1) < 0$ cannot be used to spatially confine molecules with static fields, because it is impossible to construct a local E-field maximum.

The significant progress gained over the years in trapping and manipulating neutral atoms with static B-fields [12] provides important enabling technologies for the experimental success of cold atom physics research. A variety of B-field traps have been implemented for various applications [3, 12]. Despite the analogy described earlier of the B-field confinement of magnetic dipoles with the E-field confinement of electric dipoles (of polar molecules), experimental efforts at trapping and manipulating polar molecules with electrostatic E-fields have become an active research topic only in recent years with the interest in cold atoms broadening into cold molecules. Meijer’s group pioneered the research in cooling and trapping of molecules with (3D quadrupole) E-fields [13]. They also demonstrated the guiding of cold molecules with a torus-shaped (2D) E-field hexapole [14]. With suitable modifications, an analogous IPT of the E-field can be constructed for polar molecules [3, 12, 15, 16].

In figure 1, we show the calculated de Stark shift for the $M_J = 1$ state of the heteronuclear molecule KRb that is connected to the $J = 2$ manifold when the E-field is absent. We adopt the same parameters as from its ground state $X \Sigma_v = 0$ as measured recently [1] with the electric dipole moment $D = 1.36 \times 10^4$ Hz m V$^{-1}$ and the rotational constant $B = 1.1139$ GHz. The calculation is accomplished by a numerical diagonalization of the Hamiltonian over a truncated basis of common eigenstates for operators $\mathbf{I}_x$ and $\mathbf{J}_z$. The diagonalization is carried out in the subspace of a conserved $M_J$ [10], and the procedure is found to be rapidly convergent. At the
Figure 1. The Stark shift of the $M_J = 1$ state for the polar molecule KRb in the electronic ground state $X^1\Sigma_\nu = 0$, for realistic electrostatic E-trap field strength [15, 16].

E-field strength we consider, the perturbation result $\psi_{M_J}$ in equation (5) with $M_J = 1$ and $J = 2$ is found to consist of an excellent approximation. This particular state, as illustrated in figure 1, is clearly a weak field seeking state. The maximum trap height can become as high as 173 MHz.

3. Rotational transformation of the wave function $|J, M_J\rangle$

Before we extend the Isoshima et al protocol [4] from B-field trapped atoms to E-field trapped polar molecules, we will briefly review the rotational properties of the spherical harmonics $|J, M_J\rangle$. Under a rotation along a unit vector $\hat{n}$ by an angle $\theta$, the wave function changes to

$$ R(\theta \hat{n})|J, M\rangle = e^{-i \hat{n} \cdot J} |J, M\rangle. $$

Expressed in terms of the Euler angles, the most general 3D rotation takes the following form:

$$ R(\alpha, \beta, \gamma) = \exp(-i \alpha J_z) \exp(-i \beta J_y) \exp(-i \gamma J_z). $$

According to the representation theory of rotation, we find that

$$ R(\alpha, \beta, \gamma)|J, M\rangle = \sum_{M'} D^J_{M'M}(\alpha, \beta, \gamma)|J, M'\rangle $$

$$ = \sum_{M'} \exp[-i M' \alpha] d^J_{M'M}(\beta) \exp[-i M' \gamma]|J, M'\rangle, $$

where

$$ d^J_{M'M}(\beta) = \langle J, M' | \exp[-i \beta J_y] | J, M \rangle $$

$$ = [(J + M)!(J - M)!(J + M')!(J - M')]^{1/2} \times \sum_{\nu} \left[ (-1)^\nu (J - M' - \nu)! (J - \nu)! (\nu + M' - M)! \nu! \right]^{-1} $$

$$ \times \left( \cos \frac{\beta}{2} \right)^{2J + M' - 2\nu} \left( -\sin \frac{\beta}{2} \right)^{M' - M + 2\nu}. $$

New Journal of Physics 11 (2009) 055019 (http://www.njp.org/)
This gives
\begin{align}
(J - M' + \nu)! &= \left[ -\frac{1}{2}(M' + M) \right]!, \quad (11) \\
(J + M - \nu)! &= \left[ \frac{1}{2}(M' + M) \right]!. \quad (12)
\end{align}

In order to assure that both \( J - M' - \nu \) and \( J + M - \nu \) are non-negative, we need to make \( M' + M = 0 \). As a result we find
\[
d_{M'M}^{J} (\pi) = (-1)^{J - M'} \delta_{M', -M'}, \quad (13)
\]
which gives
\[
R(\alpha, \pi, \gamma | J, M) = (-1)^{J+M} \exp[iM\alpha] \exp[-iM\gamma] | J, -M \rangle. \quad (14)
\]

We want to stress at this point that the angles \( \alpha, \beta \) and \( \gamma \) are parameters used to specify arbitrary rotations, and they have nothing to do with the internal state angles \( \theta, \phi \). To understand the physics behind the vortex generation protocol, which is directly related to the geometrical properties of the static E-field, we first consider several special cases of unit vectors as rotation axes: for \( \hat{n} = \hat{X} \cos \phi + \hat{Y} \sin \phi, \hat{n}_{\perp} = -\hat{X} \sin \phi + \hat{Y} \cos \phi \) and \( \hat{n}_{q} = \hat{X} \sin \phi + \hat{Y} \cos \phi \), respectively, with \( \hat{X} \) and \( \hat{Y} \) being the unit vectors along the Cartesian \( x \)- and \( y \)-directions. \( \theta \) and \( \phi \) denote the polar and azimuthal angles of the center of mass coordinate \( \hat{R} \). The last case of \( \hat{n}_{q} \) corresponds simply to the vertical direction of an IPT E-field in the plane of \( Z = 0 \). Using the relationship derived above, we find that
\[
\begin{align}
R_{\hat{n}}(\pi | J, M_{J}) &= \exp(-i\pi \hat{n} \cdot \hat{J}) | J, M_{J} \rangle \\
&= R_{\hat{z}}(\phi - \pi/2) R_{\hat{y}}(\pi/2 - \phi) | J, M_{J} \rangle \\
&= - (1)^{J+M_{J}} \exp[2iM_{J}\phi] | J, -M_{J} \rangle, \quad (15)
\end{align}
\]
\[
\begin{align}
R_{\hat{n}_{\perp}}(\pi | J, M_{J}) &= \exp(-i\pi \hat{n}_{\perp} \cdot \hat{J}) | J, M_{J} \rangle \\
&= R_{\hat{z}}((\pi/2 + \phi) - \pi/2) R_{\hat{y}}(\pi/2 - (\pi/2 + \phi)) | J, M_{J} \rangle \\
&= (1)^{J+M_{J}} \exp[2iM_{J}\phi] | J, -M_{J} \rangle, \quad (16)
\end{align}
\]
\[
\begin{align}
R_{\hat{n}_{q}}(\pi | J, M_{J}) &= \exp(-i\pi \hat{n}_{q} \cdot \hat{J}) | J, M_{J} \rangle \\
&= R_{\hat{z}}((\pi/2 - \phi) - \pi/2) R_{\hat{y}}(\pi/2 - (\pi/2 - \phi)) | J, M_{J} \rangle \\
&= (1)^{J+M_{J}} \exp[-2iM_{J}\phi] | J, -M_{J} \rangle. \quad (17)
\end{align}
\]

These properties of rotational transformation show that by enforcing a rotation of the internal state, the wave function gains an appropriate topologically phase specified by the azimuthal angle coordinate of the center of mass (or the molecule) coordinate. Provided the internal state flipping of the permanent dipole moment and the center of mass motion are both adiabatic, the above results show that different vortical phases are generated as in the protocol of Isoshima et al for B-field trapped atomic spinor condensates [4]–[6].
4. Vortex creation in a condensate of heteronuclear molecules

In this section, we will confirm numerically the vortex creation protocol for a condensate of heteronuclear molecules in an E-field IPT. Basically, we will simulate the axial E-field bias flip and check for the adiabatic conditions of the associated internal state. There are two main points that we need to watch for. Firstly, we need to create the proper vortical phase structure on a condensate during the time evolution of the E-field. Secondly, we must maintain adiabaticity during the flip of the internal state of a polar molecule.

Considering the first question, assume that the initial state corresponds to a polar molecule locally aligned along the E-field direction of an E-field IPT, which is essentially along the axial z-axis direction close to the z-axis, its local internal state \( \psi_{M_J}(\theta, \phi) \) can then be described approximately as the eigenstate of the system in the local E-field. Since \( M_J \) is a conserved quantity, this eigenstate \( \psi_{M_J}(\theta, \phi) \) can be expanded by a series of \( |J, M_J\rangle \) in \( J \),

\[
\psi_{M_J}(\theta, \phi) = \sum_J C_J |J, M_J\rangle. \tag{18}
\]

The flipping of the E-field bias adiabatically corresponds then to nothing but a rotation of the initial state along the unit vector \( \hat{n}_q(\theta, \phi) \) in the transverse plane by an angle of \( \pi \). According to the rotation properties we discussed before, this flipping of the bias then gives

\[
R_{\hat{n}_q}(\pi) \psi_{M_J}(\theta, \phi) = \sum_J (-1)^{J+M_J} C_J \exp[-2iM_J \phi] |J, -M_J\rangle, \tag{19}
\]

which gains clearly a vortex with a winding number \(-2M_J\).

Turning to the second question concerning the adiabaticity, we simply can propagate the initial wave function \( \psi_{M_J} \) in real time, simulating the complete process of the bias flip. To test the level of adiabaticity and the validity of equation (19), we consider points \((X_0, Y_0, 0)\) along a circle in the \(X-Y\)-plane. The absolute value for the E-field along the circle is then a constant, although their local directions are all different. In fact, for the E-field IPT being considered here, the E-field can be written as \( \vec{E} = E'(X, -Y, L) \) where \( L \) is a constant. Choosing the weak field seeking state \( \psi_{M_J=1} \) with \( J = 2 \) at \( \vec{E} = 3 \text{ kV cm}^{-1} \hat{Z} \) as considered in figure 1, we simulate the flip protocol over various time intervals. The time evolution of the bias E-field is taken to be the same as that in the optimal B-field protocol [9], and the E-field in the transverse \( X-Y\)-plane is treated as a constant.

In our simulation, we take the E-field at \( t = 0 \) as \( E'L = 3 \text{ kV cm}^{-1} \) and \( E' \sqrt{X_0^2 + Y_0^2} = 0.3 \text{ kV cm}^{-1} \). After the flip of the E-bias over \( 10^5/B \) (\( \sim 0.1 \text{ ms at } B = 1.1139 \text{ GHz} \)), which is a reasonably fast timescale, we calculate the inner product of the final state with the eigenstate \( \psi_{M_{J-1}} \) (of \( J = 2 \)) when the E-field \( \vec{E} = -3 \text{ kV cm}^{-1} \hat{Z} \) is pointed in the downward direction. Using these parameters, the adiabaticity is found to be fully satisfied, and the state overlap is essentially 100% except for the extra topological phase gained as shown in figure 2. A numerical fit gives precisely the slope for the phase over \( \phi \) being exactly equal to \(-2\), which confirms the high fidelity operation of our vortex pump proposal.

Before concluding, we point out that, like the system of magnetically trapped atomic spinors inside a B-field IPT [9, 17], the quantity of \( J - L_z \) is found to commute with the Hamiltonian \( BJ^2 - D \cdot \nabla X'(-X, -Y, L) \), where \( E' \) is the spatial gradient of the E-field IPT and \( L_z \) is the mechanical angular momentum of the heteronuclear molecule. \( \vec{J} \) is the rotational angular momentum of the molecule. If the flip of the E-bias is indeed adiabatic, the respective quantum
Figure 2. The topological phase gained after numerically flipping the bias E-field, which is calculated by taking the inner product of the time evolved state after flip with the nominally down polarized ($M_J = -1$) state. The closed squares denote numerically computed data, and the red solid curve represents a linear fit. numbers conserve the combination $M_J - L_z$. After going through the flip, the internal rotational state is changed from $M_J$ to $-M_J$, which then must be accompanied by an increase of $2M_J$ to its mechanical angular momentum. Thus we see the appearance of a vortex state.

In conclusion, by analogy with B-field trapping of neutral atoms with magnetic dipoles, we study and identify weak field trapping states of polar molecules inside a spatially inhomogeneous dc E-field. Further, we suggest that an effective and efficient vortex pump protocol can be envisioned for condensates of polar molecules [8, 9], based on the flipping of the axial bias field, originally suggested [4] and experimentally demonstrated for atomic spinor condensates inside a B-field IPT [5]–[7]. We have confirmed that the E-field bias flip remains effective, and the vortex state created maintains a vorticity proportional to the $M_J$ quantum number of the trapped molecule state. When a diatomic molecule is placed inside a homogeneous E-field, $M_J$ is a good quantum number. The weak field trapping states can be associated with very large values of $M_J$, thus the amount of vorticity created could become very significant even after a single bias flip. This vorticity possibly could open up a practical approach to reach the rapid rotation limit of atomic/molecular quantum gases.

More generally, we find that this protocol for vortex creation remains effective if the diatomic molecule is taken as a symmetric top [18]. Interestingly, we find that the E-bias flip also works for the strong field trapping states provided the polar molecules are confined through other means not relying on its permanent electric dipole. Finally, other improvements to the B-field bias flip protocol [4], such as those developed for cyclically operated continuous vortex pumping schemes, can be analogously extended to the case of heteronuclear molecules [8, 9].

Acknowledgment

This work was supported by the US NSF, the NSF of China under grant no. 10640420151 and the NKBRSF of China under grants nos. 2006CB921206 and 2006AA06Z104.
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