Rotating molecules in optical lattices, alignment and monopole crystals

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The recent progress towards production of near-ground state quantum-degenerate molecules raises the issue of how such “small” molecules behave in an optical lattice. In this Letter we show that the coupling of the molecular orientation to the local electric field direction will provide several new phenomena. In the case where the lasers forming different crystallographic directions of the lattice are incoherent, the orientation of the molecules is conserved (for $L = 1$) and a novel form of anisotropic superfluidity can be expected. When the lasers are coherent, and the optical lattice is such that the splitting of the rotational levels is large compared to the centre of mass energies, an adiabatic description of the molecular orientation is appropriate. This leads to geometric vector potentials, pseudo-magnetic monopoles and a frustrated band structure with degenerate minima.

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The rapid progress in making molecules in ultracold atomic gases using Feshbach resonances\cite{1} and more recently using photoassociation\cite{2,3} has provided a new direction for degenerate gas physics, both in terms of non-equilibrium effects due to Feshbach sweeps of magnetic fields but also equilibrium many body phenomena such as the BEC-BCS crossover. The first non-s wave molecules have been studied in Cs$_2$\cite{4} and very recently in K$_2$\cite{5}. Novel phenomena such as splitting of the different multiplet Feshbach resonances has been observed. It was soon realised that optical lattices could provide a very sheltered environment for the production of molecules due to control of occupancy (leading to a lack of harmful three-body collisions). Initially this led to photoassociative state-selective production\cite{6}. More recently the combination of the tunability of the optical lattice in terms of tunneling (through laser intensity) and photoassociative state-selective production\cite{6} has provided a new direction for degenerate gas physics, both in terms of incoherent\cite{3} and bosons\cite{5}. The reliability of the occupancy in the lattice of the molecules is now very good\cite{9}.

Very recently Raman down steps (STIRAP)\cite{10} towards ground state molecules have been achieved and with the proposal of frequency-comb methods\cite{8}, it is timely to consider the nature of near-ground state molecules in optical lattices.

Large molecules (i.e. with an extent large compared to the optical lattice parameter) have been studied theoretically in optical lattices by Fedichev et al\cite{11}. The nature of binding on scales large compared to the optical lattice parameter were established in some detail. For such a large s-wave molecule there is no coupling to the orientation of the lattice except through the anisotropy of the center of mass motion.

In this Letter we consider small molecules where there is a significant orientational coupling to the optical lattice due to their inherently anisotropic polarisability. We will see that this coupling has several pronounced effects on the centre of mass motion, raising issues of the nature of resulting condensed states. (For larger molecules, this presumably corresponds to interference between the incident optical field and light scattered by one of the constituent atoms, as experienced by the other atom.)

The most striking effects are in an excited rotational state which might cause disquiet in terms of lifetimes of such states. However we take comfort from the case of ortho- and para-Hydrogen\cite{14,15}, where ortho-Hydrogen (which has $L = 1$) has a lifetime at condensed matter densities of the order of a day due to the very small matrix elements for relaxation of the nuclear $I^{\text{tot}} = 1$ triplet to the singlet.

In fields of the strength\cite{12,13} occurring in optical lattices, $L^2$ is a good constant of the motion despite the manifest lack of spherical symmetry. This is due to the rotational level spacing being typically six orders of magnitude larger than the recoil energy. However multiplet splitting (as against mixing) already indicates coupling of the orientation to the local field direction and it is the consequences of such a coupling in an optical lattice that we will explore in this Letter.

Let the molecule have polarisabilities $\alpha^\parallel$ and $\alpha^\perp$ parallel and perpendicular to the molecular axis respectively, and define the average polarisability, $\bar{\alpha} = \frac{1}{2}(\alpha^\parallel + 2\alpha^\perp)$, and $\delta\alpha = \alpha^\parallel - \alpha^\perp$. We assume from now on that the lattice is red-detuned and that $\delta\alpha > 0$. Then to second order in the applied field, $E(z)$, the contribution to the Hamiltonian, $H^{(l)}$, for a given multiplet is:

$$H^{(l)} = -\frac{1}{8}\vec{E}^2 \left( \bar{\alpha} - \frac{3}{2}\delta\alpha \sum_{m=-l}^{l} \frac{3m^2 - l(l+1)}{(2l+3)(2l+1)} |m\rangle \langle m| \right)$$

(see, for example ref\cite{16}) where the angular momentum quantum numbers are $l$ and $m$ with the axis of quantisation parallel to the field.

For a one-dimensional position-dependent electric field, $E(X)\vec{z}$, where $X$ is the centre of mass position of the molecule, we define (now considering $L = 1$):

$$H_{z}(X) = -\frac{1}{8}\vec{E}^2(X) (\alpha_1 + \alpha_2) |m_z = 0\rangle \langle m_z = 0|$$

where $z$ denotes the direction of the field, $\alpha_1 = \bar{\alpha} -
\[ \frac{1}{\hbar} \delta \alpha \quad \text{and} \quad \alpha_2 = \frac{1}{\hbar} \delta \alpha. \] The physical interpretation of this is semiclassical. For the states \( m = \pm 1 \) the molecule presents its less polarisable directions to the field. This means the barriers to tunneling are reduced compared to random orientation but also the minima are raised compared to random orientation. \( m = 0 \) presents (semiclassically) the most polarisable axis half the time and hence has higher barriers, but lower minima.

We see that in one dimension the orientation of the molecule is a constant of the motion. It is perhaps more surprising that we can make statements of a similar nature in higher dimensions. However the mutual coherence or incoherence of the lasers in the different axes of the optical lattice affects the results substantially.

In the incoherent case, let the polarisation of the electric field of the laser associated with optical lattice axes \( i = x, y \) or \( z \) be denoted by the unit vectors \( \hat{e}_i \). Then the perturbed energy is the sum of three terms of the form [2]:

\[
\mathcal{H}(\mathbf{R}) = \mathcal{H}_{\hat{e}_x}(X) + \mathcal{H}_{\hat{e}_y}(Y) + \mathcal{H}_{\hat{e}_z}(Z) = -\frac{\hbar}{\lambda} \sum_{i=x,y,z} \mathcal{E}^2(R_i) (\alpha_1 + \alpha_2)|m_i = 0\rangle\langle m_i = 0| \tag{3}
\]

where \( m_i \) is an eigenvalue of \( \hat{e}_i \cdot \mathbf{L} \).

The result is particularly straightforward if the set \( \{\hat{e}_i\} \) is orthogonal. Then the set \( \{m_i = 0\} \) (the “polar basis”) is also orthogonal. In that case the three contributions to the Hamiltonian from the projection operators \( |m_i = 0\rangle\langle m_i = 0| \) commute and hence the Hamiltonian is diagonal. A molecule in one of the eigenstates, \( |m_i = 0\rangle \), will experience two directions with small periodic potentials in the direction parallel to \( \hat{e}_i \), a deeper one. For \( L > 1 \), the situation is more complicated: for example it is not guaranteed that the \( m = 0 \) states along three axes are orthogonal (e.g. \( L = 2 \)), so the projection operators do not commute.

The basis set allows some translation of the \( S = 1 \) spinor results from references [13]. The molecule-molecule scattering will have singlet and quintuplet scattering lengths which determine whether the ground state is a polar state (as in the \( S = 1 \) case) which is compatible with the diagonalised Hamiltonian, albeit with anisotropic superfluid properties. It is less clear what happens for the case which would be ferromagnetic without the lattice.

Let us now turn to the coherent case. Here the polarisation of the optical lattice is position-dependent. For definiteness, pick

\[
\mathbf{E}(\mathbf{R}) = \mathcal{E}_0 \left( \hat{x} \sin kZ + \hat{y} \sin kX + \hat{z} \sin kY \right) \cos \Omega \tag{4}
\]

and for future reference define \( \mathcal{E}(\mathbf{R}) = \mathcal{E}(\mathbf{R}) \). Then the antinodes, i.e. the minima of the optical lattice potential, are at \( \mathbf{R} = (\lambda/4)(2\ell + 1, 2m + 1, 2n + 1) \). There are four distinct minima (as the Hamiltonian does not distinguish the sign of the electric field), due to the relative signs of the components of \( \mathbf{E} \).

The case where there is an adiabatic separation of the energy scales of splitting the angular momentum multiplet and the centre of mass motion in one of the optical lattice wells (and hence in an even more pronounced form in terms of tunneling) will be the focus for the remaining part of this Letter. In that limit the molecule preserves its orientational state relative to the local electric field direction, determined by the centre of mass position. Unlike the incoherent case the results are qualitatively similar for all \( L > 0 \). Berry and Robbins [17] studied a related problem where there was a linear coupling between angular momentum and spatial variables in an adiabatic limit which we will comment on presently.

We form the adiabatic basis, \( |m, \mathbf{R}\rangle \), by rotating the reference basis set, \( |m\rangle \), so that the \( z \)-axis is transformed to the local electric field direction appropriate to centre of mass position \( \mathbf{R} \):

\[
|m, \mathbf{R}\rangle = D(-\eta(\mathbf{R}), -\zeta(\mathbf{R}), 0)|m\rangle = \exp(i\eta(\mathbf{R})\sigma_z) \exp(i\zeta(\mathbf{R})\sigma_y)|m\rangle
\]

where \( \{\sigma_i\} \), with \( i = x, y \) or \( z \), are the spin-1 Pauli matrices. Thus

\[
\hat{E}(\mathbf{R}) \cdot \mathbf{L}|m, \mathbf{R}\rangle = m|m, \mathbf{R}\rangle
\]

with \( \hat{E}(\mathbf{R}) \) is the unit vector corresponding to the local electric field.

We then look for an approximation to the solution, \( \Psi(\mathbf{R}, \zeta, \eta) \), of the Schrödinger equation for the molecule

\[
\left[ -\frac{\hbar^2}{2M} \nabla^2_{\mathbf{R}} - \frac{1}{\hbar^2} \mathcal{E}^2(\mathbf{R}) (\alpha_1 + \alpha_2)|0, \mathbf{R}\rangle\langle 0, \mathbf{R}| \right] \Psi = \epsilon \Psi
\]

The adiabatic approximation is to assume

\[
\Psi(\mathbf{R}, \zeta, \eta) \simeq \sum_{m = -1}^{1} \psi_m(\mathbf{R}) |m, \mathbf{R}\rangle
\]

which leads (upon substitution and performing an integration over the angular variables) to the effective Schrödinger equation in terms of a vector potential [20]

\[
\mathbf{A}^{nl} = -i\langle n, \mathbf{R}| \nabla \mathbf{R}|l, \mathbf{R}\rangle :
\]

\[
\left( -\frac{\hbar^2}{2m} \Delta^{nl} \cdot \Delta^{lm} + V^{nm}(\mathbf{R}) \right) \psi^{nl}(\mathbf{R}) = E \psi^{nl}(\mathbf{R}) \tag{5}
\]

where we define the covariant derivative, \( \Delta^{nl} = \delta^{nl} \nabla_{\mathbf{R}} - i\mathbf{A}^{nl} \) and

\[
V^{nm}(\mathbf{R}) = -\frac{1}{\hbar^2} \mathcal{E}^2(\mathbf{R}) (\alpha_1 \delta^{nm} + \alpha_2 P_0^{nm}) + \sum_{l \neq n} \mathbf{A}^{nl} \cdot \mathbf{A}^{lm} \delta^{nm}
\]

(with no summation convention), where \( P_0^{nm} = \delta^{n0} \delta^{n0} \). Non-Abelian vector potentials lead to effects experienced...
by multi-level atoms in light fields: the case of spin-orbit effects, laser-assisted tunneling and tripod-field induced degenerate dark states. This Letter contains the first example of the new phenomena experienced in a three-dimensional extended setting with such non-Abelian fields.

We may evaluate the vector potential:

$$A^{nl} = \left[ (\sigma^x z \cos \zeta + \sigma^y z \sin \zeta) \nabla R \eta + \sigma^y \nabla R \zeta \right]$$

and hence determine the associated field:

$$F^{nn}_{ij} = \partial_i A^{mn}_j - \partial_j A^{nm}_i - i[A_i, A^j]^{mn} = 2(\partial_i \eta \partial_j \zeta - \partial_j \eta \partial_i \zeta)(\sigma^x_{z} \cos \zeta - \sigma^y_{z} \sin \zeta)$$

implying that the pseudo-field, $B^{nn}_i = \frac{1}{2} \epsilon_{ijk} F^{mn}_{jk}$ is

$$B^{nn} = 2 \nabla \zeta \times \nabla \eta (\sigma^x_{z} \cos \zeta - \sigma^y_{z} \sin \zeta)$$

with the polar angles, $\zeta$ and $\eta$, playing the role of “Euler potentials” for the field $B$.

An immediate consequence of equation (6) is that $\nabla \cdot B = 0$ except possibly at singularities of $\nabla \eta$ and/or $\nabla \zeta$, which occur at the nodes of the electric field of the optical lattice. Firstly consider the form of the electric field in the vicinity of the node at the origin

$$E(R) \propto k E (\hat{\xi} Z + \hat{\eta} X + \hat{\zeta} Y)$$

To examine the nature of the pseudomagnetic field and its flux, it is easiest to calculate $\nabla \times E$ in polar coordinates around the node. The relation between the polar coordinates of the electric field vector, $(\zeta, \eta)$, and those of the centre of mass $(\theta, \phi)$ are determined implicitly from:

$$\hat{E}_x = \sin \zeta \cos \eta = Z/R = \cos \theta$$
$$\hat{E}_y = \sin \zeta \sin \eta = X/R = \sin \theta \cos \phi$$
$$\hat{E}_z = \cos \zeta = Y/R = \sin \theta \sin \phi$$

Using expressions for $\cos \zeta$ and $\tan \eta$ we find after some algebra that:

$$\nabla \zeta \times \nabla \eta = -\frac{\hat{R}}{R^2} \frac{1}{\sin \zeta}$$

and hence

$$B^{nn}(R) = \frac{\hat{R}}{R^2} (\sigma^x_{z} - \sigma^y_{z} \cot \zeta)$$

in the vicinity of the node.

We will now calculate the flux through a spherical surface of $B^{nn}$. We will neglect the Dirac-like strings which emerge along the positive and negative $y$-axes (reminiscent of the two strings in Schwinger’s treatment of monopoles), as the strings are irrelevant to our final lattice treatment.

The second term in Eqn. (7) vanishes as there is a cancelation from the contributions from $0 \leq \phi < \pi$ and $\pi \leq \phi < 2\pi$. The first term provides $8\pi^2 \sigma_2$ flux through the surface, corresponding to monopoles of charge $\pm 8\pi$. These are the analogues of the monopole in the the work of Berry and Robbins.

The field in the vicinity of the nodes at $R = (m, n, \ell)$ has the form:

$$E(R) \propto (-1)^\ell (Z - \ell) \hat{\xi} + (-1)^m (X - m) \hat{\eta} + (-1)^n (Y - n) \hat{\zeta}$$

In the case of one or three of $\ell$, $m$ and $n$ being odd, then either the sign of $\nabla \eta$ or $\nabla \zeta$ reversed. Thus the $(m = 1)$ monopole has negative charge. Conversely, if there are zero or two odd integers, the $(m = 1)$ monopole has a positive charge as neither or both gradients are reversed.

Thus the monopoles reside in a Na-Cl lattice, coincident with the nodal points of the electric field, of alternating positive and negative magnetic charges. The signs of all charges are reversed if the angular momentum of the molecule is reversed.

Since the adiabatic limit will require a strong optical lattice it is natural to construct the tight binding model corresponding to the continuum Schrodinger equation, Eqn. (4). The lattice is cubic and in the adiabatic limit the distinction between the field directions on the different sites in the unit cell is irrelevant as they are all perfectly adiabatically connected and degenerate in energy. The monopoles reside on the dual lattice, with the opposite signs adopting a Na-Cl structure. We will focus on the $m = 1$ case.

We must assign Peierls factors, $e^{iA_{n,n'}}$, to the hopping term on each link between sites $n$ and $n'$. The flux, $\Phi_{n,n',n''n'''},$ through a plaquette is counted modulo $2\pi$ as $e^{i\Phi} = e^{\imath \sum \Delta A}$. Then we must pick $A_{n,n'}$ so that we get a flux of $\pm 2\pi/3$ through each plaquette, so we obtain a total flux out of the six plaquettes surrounding a monopole of $\pm 8\pi$. The Peterls factors are chosen as indicated in Fig. (1), where we see there are two sites per unit cell denoted $A$ and $B$. This leads to the secular determinant (here $t > 0$ and $\omega = e^{i\pi/3}$):

$$\begin{vmatrix}
-\epsilon & -2t(\omega X + Y + \omega^{-1} Z) \\
-2t(\omega^{-1} X + Y + \omega Z) & -\epsilon
\end{vmatrix} = 0$$

where: $X = \cos k_x$ and similarly for $Y$ and $Z$. This implies

$$\epsilon_{\pm}(k) = \pm \sqrt{2} t \sqrt{(X - Y)^2 + (Y - Z)^2 + (Z - X)^2}$$

The two bands touch along the $(\pm 1, \pm 1, \pm 1)$ directions, with the dispersion relation in that vicinity (defining $k^\parallel$ parallel to the $(1,1,1)$ direction and $k^\perp$ ($\geq 0$) radially perpendicular to the direction) being $\epsilon_{\pm}(k^\parallel, k^\perp) \simeq \pm \sqrt{3} \sin(k^\parallel / \sqrt{3})k^\perp$ for $k^\perp / k^\parallel < 1$.

The most important aspect of the dispersion relations relates to the frustrated nature of the Hamiltonian. The minima of $\epsilon_{-}(k)$ are at the three points in the Brillouin Zone: $k = (0,0,\pi)$ and cubic equivalents. Because the
and a simple cubic system. The wavefunctions on the energy are outside the first Brillouin Zone and equivalent to the FCC reciprocal lattice. Thus the minima in energy are $-4t$ which is higher than the $-6t$ expected of a simple cubic system. The wavefunctions on the $A$ and $B$ sublattices, $(\psi_A, \psi_B)$, for the three minima are: for $k(Z) = (0, 0, \pi)$, $(\psi_A^Z, \psi_B^Z) = (1, -\omega)$; for $k(Y) = (0, \pi, 0)$, $(\psi_A^Y, \psi_B^Y) = (1, 1)$; for $k(X) = (\pi, 0, 0)$, $(\psi_A^X, \psi_B^X) = (1, -\omega)$.

The three minima, in conjunction with the possible competition of singlet and quintuplet scattering lengths, make the nature of the mean field ground state in general unclear.

In conclusion we have shown that in both incoherent and coherent optical lattices the orientation of a molecule with $L \neq 0$ couples to the direction of the optical field. In the incoherent case the orientation is conserved in a highly symmetric manner for $L = 1$. For the coherent case the strong optical fields lead to the angular momentum around the local electric field being an adiabatic invariant. The consequences of the adiabatic approximation failing at the nodal points of the lattice, in the classically disallowed region, affects the tunneling of the molecule. This leads to pseudomagnetic monopoles with their flux influencing the band structure of the molecule in a striking manner. The consequences for condensation remain to be explored, as does the configuration of monopoles in cases of general (eg non-bipartite) dual lattices.

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