Optically induced conical intersections in traps for ultracold atoms and molecules

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We show that conical intersections can be created in laboratory coordinates by dressing a parabolic trap for ultracold atoms or molecules with a combination of optical and static magnetic fields. The resulting ring trap can support single-particle states with half-integer rotational quantization and many-particle states with persistent flow. Two well-separated atomic or molecular states are brought into near-resonance by an optical field and tuned across each other with an inhomogeneous magnetic field. Conical intersections occur at the nodes in the optical field.

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There is great interest in the properties of ultracold atoms in traps with unusual shapes, especially in rings and other multiply connected geometries. Both magnetic [1–4] and optical [5] ring traps have been created. Such traps have potential uses in inertial sensing and in atom interferometry. They also exhibit geometric phases (Berry phases) due to the spatial variation in the magnetic field direction [6–8]. These quantum phases may be exploited to create neutral-atom analogues of systems involving charged particles in magnetic fields [7]. Lin *et al.* [9] have recently used Berry phases generated with a spatially varying Raman coupling to create large synthetic magnetic fields, with potential for studying phenomena such as the quantum Hall effect and topological quantum computing.

Berry phases of a qualitatively different type would be be accessible in a ring trap with a conical intersection (CI) at the center. A CI occurs when two adiabatic potential energy surfaces intersect, at a point in two dimensions or on a surface of dimension $(n-1)$ in n dimensions. The surfaces have the topology of a double cone near the intersection. A CI produces a *half-integer* Berry phase [10, 11]: the internal wavefunction of a state that fully encircles the intersection adiabatically changes sign upon completion of a circuit around it.

CIs can exist in the internal vibrational coordinates of polyatomic molecules, [12], where they have important consequences for collision dynamics and photochemistry, and in momentum space, where they have important consequences for electronic band structure and conductivity. CIs in momentum space are often referred to as *Dirac cones*, and can occur both in solid-state systems such as graphene [13, 14] and in cold atoms in specific laser and optical lattice configurations [15–17]. Moiseyev *et al.* [18] have shown that arrays of CIs may occur in a combination of molecular rovibrational coordinates and spatial coordinates for diatomic molecules in optical lattices, and may have important consequences for the dynamics of ultracold molecules in standing laser waves. Wüster *et al.* [19] have proposed creating CIs with groups of ultracold atoms or molecules interacting through resonant dipole interactions.

We have recently shown that, for a gas of ultracold molecules with both electric and magnetic dipoles, CIs can be created purely in laboratory position space with externally applied static electric and magnetic fields [20]. The magnetic field is used to produce a crossing between two states of opposite parity, and the electric field breaks the parity symmetry and induces an avoided crossing between the states [21, 22]. An CI is created where the electric field passes through zero. If the intersection is surrounded by an optical dipole trap, the resulting adiabatic potential has a toroidal minimum that encircles the CI. The geometric phase effect can then produce a ground state with persistent flow and half-integer quantization of the angular momentum for rotation around the ring.

CIs of the type described in ref. [20] are unlikely to be accessible for atoms, because there are no coolable atomic systems that have two states of opposite parity close enough together to be brought into degeneracy by a magnetic field. However, a stable degenerate gas of molecules with both electric and magnetic dipole moments is also some way away. The object of the present paper is to propose an alternative that is feasible for atoms, in which two states that cross as a function of magnetic field are coupled by a microwave or laser field. For generality, we refer to the microwave or laser field as an optical field. The coupling is proportional to the amplitude of the optical field, so CIs occur at nodes in the field, where the amplitude is zero. This approach has the advantage that the two states to be coupled do not need to be near-degenerate before the optical field is applied. In addition, it allows CIs to be created between pairs of states of the *same* parity, which are coupled by the magnetic component of the optical field.

CIs of the type proposed here will make it possible to study new types of interference effect in ultracold atomic systems. In addition to the half-integer quantization and persistent flow discussed in ref. [20], it will be possible to explore dynamical effects produced by the Berry phase. For example, if portions of a matter wave pass either side of the CI, the two parts will interfere destructively when they meet on the far side. If two or more CIs can be produced in the same trap, as described below, even richer interferences will be possible between paths that encircle one CI and change sign and paths that encircle zero or two CIs and do not.

We consider a two-level atomic or molecular system with ground and excited states $|g\rangle$ and $|e\rangle$, with zero-field energy separation $\hbar\omega_0$, coupled by a standing-wave optical field with frequency $\omega_{\rm L}/2\pi$ and a node at $y = 0$. An inhomogeneous magnetic field $B_z(x)$ is oriented along the laboratory z axis and varies along the x axis. The dressed-state Hamiltonian is

$$\begin{pmatrix} \hbar\omega_{\rm L} + \mu_g B_z(x) & (\hbar\Omega_{eg}/2)\sin ky \\ (\hbar\Omega_{eg}/2)\sin ky & \hbar\omega_0 + \mu_e B_z(x) \end{pmatrix}, \qquad (1)$$

where Ω_{eg} is the Rabi frequency of the transition and $k = \omega_{\rm L}/c$. If the optical field is resonant with the transition between the two states at a magnetic field $B_z(x = 0) = B_0$, the eigenvalues of (1) will form a seam of CIs along a line at (0,0,z). If the atomic or molecular cloud is large enough, there will be additional seams of CIs at regular intervals such that $y_n = n\pi c/\omega_{\rm L}$ for integer n.

CIs of this type can in principle occur for any optical frequency. However, the sharpness of the excited state is limited by spontaneous emission, which is likely to limit the use of visible and even near-infrared transitions because of the ω_0^3 factor in the equation that relates the Einstein A coefficient to the transition dipole. This might be overcome for strongly forbidden transitions, but then the Rabi frequencies would be quite low. We will return to the possible use of infrared transitions below, but we will begin by considering CIs based on microwave transitions.

An alkali-metal atom with nuclear spin I has two zero-field hyperfine states with $F = I \pm \frac{1}{2}$, each of which splits into $2F + 1$ sublevels in a magnetic field. The two states with the same M but different values of F have equal and opposite Zeeman effects. There are magnetic-dipole-allowed microwave transitions between states (F, M) and $(F{+}1,M')$ with $\Delta M{=}M'{-}M{=}0,{\pm}1$. As an example, we consider ^{87}Rb, which has $I = \frac{3}{2}$ and a zero-field splitting of 6.835 GHz between its $F = 1$ and $F = 2$ states. Let us consider a central static field $B_0 = 200$ G at $y = 0$, so that the splitting between the $(F, M) = (1,{+}1)$ and $(2,{+}1)$ sublevels is 7.115 GHz. The transition between these states has $\Delta M = 0$ so requires an oscillating magnetic field along the z axis. To provide this, consider placing the atoms in a rectangular or cylindrical microwave cavity in which the transverse electric mode TE_{011} is excited. This creates magnetic field lines that circle in the yz plane, with a node in $B_z(\omega_L t)$ at $y = 0$.

To create an observable CI in a BEC, it seems reasonable to aim for a ring radius of 6 μm. This could be achieved with a magnetic field gradient dB_z/dx of 50 G/cm and an optical trap with a force constant of 28 nK/μm^2. To balance the Zeeman effect would require an optical Zeeman effect of the same magnitude, i.e. about 1 μK at $y = 6$ μm, corresponding to $\Omega_{eg} \sin ky = 21$ kHz at $y = 6$ μm or 33 MHz at the magnetic field antinode for a cubic cavity. This is a realistically achievable field, and indeed Spreeuw *et al.* [23] in 1994 demonstrated trapping of ground-state Cs atoms in a microwave trap with a central Rabi frequency of 36 MHz in a spherical cavity, despite a relatively low cavity Q-factor of 5500 due to the need for holes to allow access for atomic and laser beams.

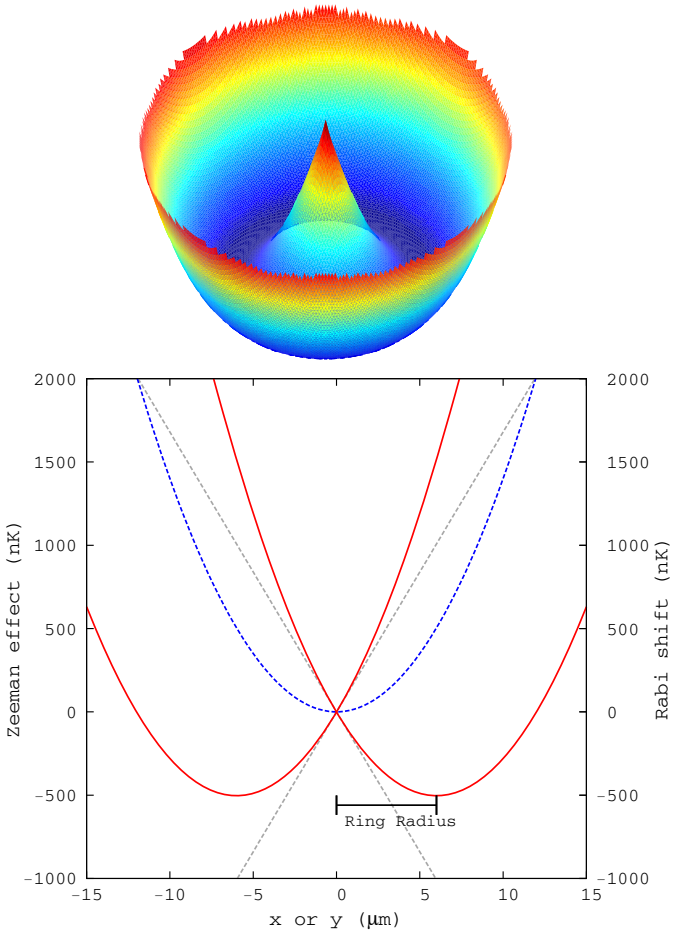

FIG. 1. (color online). Upper panel: toroidal potential formed by creating an optically induced conical intersection at the centre of an optical dipole trap; lower panel: contributions of the individual contributions to the toroidal potential for ^{87}Rb for the parameters given in the text.

Such a trap would create a potential minimum around a ring in the xy plane, about 0.5 μK below the energy of the CI, as shown in Fig. 1. Rubidium atoms are far too massive to tunnel into such a potential and reach the central point. There is no point at which the magnetic field is zero, so losses due to Majorana spin flips should not be a problem. The arrangement described thus far is cylindrically symmetrical, and confinement in the z direction must be provided optically, since the trapped atoms are in a combination of two states with different Zeeman effects and cannot be levitated magnetically.

The quantum behaviour of an atom or a BEC in a trap of this type is exactly the same as discussed by Wallis *et al.* [20] for a CI induced by static fields. For a single particle, the Berry phase due to the CI produces antiperiodic boundary conditions $\Phi(\phi + 2\pi) = -\Phi(\phi)$ for the motion around the intersection, where ϕ is the angle in the xy plane. If the ring is sufficiently flat, the Berry phase produces states with half-integer rotational angular momen-

tum around the intersection. This differs from the Berry phases that can exist in quadrupole magnetic traps or storage rings [6–8], because in our case the magnetic field does not vary in direction around the ring. For a BEC that can be modeled by the Gross-Pitaevskii equation, the self-energy is itself anisotropic because the molecular eigenstate changes from $|g\rangle$ to $|e\rangle$ around the ring. The ground state is a flowing state with half-integer angular momentum if the average self-energy is large enough to overcome its own anisotropy and that of the potential [20]. ^{87}Rb is a particularly favorable case in this respect, because its singlet and triplet scattering lengths are both positive and quite similar to one another, so that all its hyperfine states have quite similar scattering lengths.

Several variants of this arrangement can be envisaged. CIs could be created for a variety of different atoms and pairs of atomic states. For microwave transitions of alkali metal atoms, however, transitions with $\Delta M = 0$ are optimum because the two states have equal and opposite Zeeman effects, so form an untilted CI. A small difference in Zeeman effects can be compensated by an offset of the optical potential, as in the static case [20], but it will be much harder to create a flat ring if the two crossing states have substantially different Zeeman effects.

It might be possible to generate microwave fields of sufficient power on an atom chip, using microwave near-fields, such as have been used for the coherent manipulation of ultracold atoms on atom chips [24, 25]. Two parallel current-carrying wires along x, with in-phase currents at microwave frequencies, will create an oscillating magnetic field with a nodal plane in the magnetic field perpendicular to the chip $B_z(y)$ half-way between the wires, in the xz plane at $y = 0$. In this case confinement might be achieved with an optical trap created by reflecting a laser beam from the chip surface [26].

It is also conceivable that a CI might be created using radiofrequency fields to couple two atomic states of the same F but different M. In this case the requirement for nearly equal and opposite Zeeman effects is met at low fields by states with equal and opposite M values, and together with the selection rule $\Delta M = \pm 1$ this requires $M = \pm\frac{1}{2}$ for a 1-photon transition. Such states exist only for fermionic atoms. However, another possibility would be to use high-field states, with magnetic moments determined mostly by the electron spin M_S rather than the total spin M. Strong radiofrequency fields have been generated on atom chips and used to shape trap potentials [27, 28], and even to generate ring-shaped traps [3]. However, even if a node in a radiofrequency field can be positioned accurately enough, it will be challenging to produce the oscillating fields needed to form a CI within a few μm of the nodal plane.

There is a final possibility that warrants consideration, though it is further from implementation than the atomic examples considered above. If light of much shorter wavelength is used to provide the optical coupling, it is possible to envisage an *array* of CIs within a single BEC, which would allow the study of even richer interference

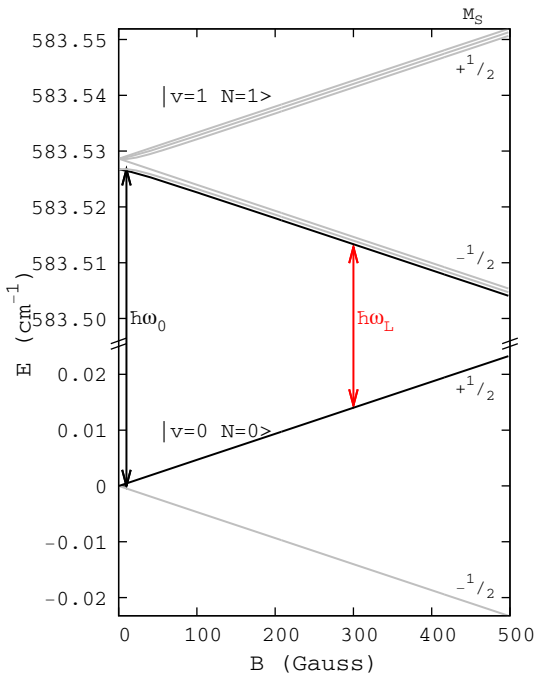

FIG. 2. (color online). Magnetic field dependence of the $|v = 0, N = 1\rangle$ and $|v = 1, N = 1\rangle$ states of CaF($X^2\Sigma^+$). To form an array of CIs the $|v = 0; N = 0, M_N = 0, M_S = \frac{1}{2}\rangle$ and $|v = 1; N = 1, M_N = 1, M_S = -\frac{1}{2}\rangle$ states (black), with a zero field separation of $\hbar\omega_0$, are coupled by a linearly polarized laser standing wave $\hbar\omega_{\rm L}$ that is resonant with the two-states at $B_0 = 300$ G.

effects. To achieve this, the spacing between intersections must be smaller than the size of the trapped ultracold gas. This requires a laser half-wavelength $\lambda_{\rm L}/2 = \pi c/\omega_{\rm L}$ less than a few hundred μm. Furthermore, it would be necessary to create an array of individual optical traps to surround each CI separately, with a controllable barrier between them. This probably requires the intersections to be separated by at least a few μm. Such traps cannot be created with the near-resonant laser that generates the CIs, but it might be achieved using a holographically generated array of microtraps [29] centered on the intersections.

An array of CIs might in principle be created for either atoms or molecules, but we are not aware of a coolable atom with a suitable transition in the required frequency range. However, the wavelength range of 3 to 300 μm corresponds to a level separation of 30 to 3000 cm^{-1}, which is typical of molecular vibrations. We therefore consider the example of CaF($X^2\Sigma^+$), which has a vibrational frequency of 588 cm^{-1} (17 μm). CaF is not yet available ultracold, though it has been produced at mK temperatures by buffer-gas cooling [30]. The levels involved in the $(v, N) = (0, 0) \leftrightarrow (1, 1)$ transition of CaF are shown in Figure 2 as a function of magnetic field. At high magnetic field all the rovibrational states are characterized by the spin-projection quantum number $M_S = \pm\frac{1}{2}$.

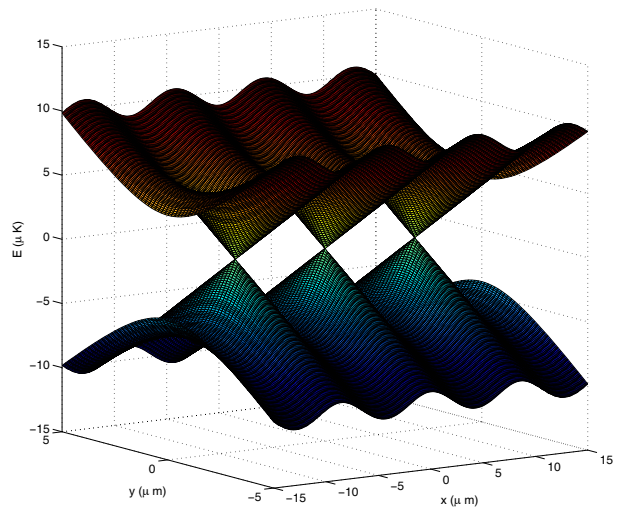

FIG. 3. (color online). Array of conical intersections formed by a 5 mW/cm^2 laser standing wave resonant with two ro-vibrational states of CaF, with a central magnetic field B_0=300 G and a constant magnetic field gradient dB_z/dy = 553 G/cm.

We consider a CI formed from the two states that are predominantly $|v = 0; N = 0, M_S = +\frac{1}{2}\rangle$ and $|v = 1; N = 1, M_N = 1, M_S = -\frac{1}{2}\rangle$, coupled by a laser linearly polarized along the magnetic field axis z. These two states are shown in black in Figure 2. The transition dipole moment between the $v = 0$ and 1 states can be estimated from the gradient of the permanent electric dipole moment of the molecule at its equilibrium bond length [31]. Assuming that the vibrational wavefunctions are harmonic, the transition dipole moment can be estimated as 0.25 D. The natural lifetime of the excited state is $\tau = 1/A_{eg} \approx 0.25$ s, so that the width due to spontaneous emission is negligible.

The dressed-state Hamiltonian is constructed with a field-free energy separation $\hbar\omega_0 = 583.5\ hc$ cm^{-1}, and a standing-wave laser with an intensity of 5 mW/cm^2, resonant at a magnetic field $B_0 = 300$ G. The intersecting dressed-state eigenvalues are shown in Figure 3. CIs occur every $\lambda_L/2 \approx 9$ μm, at each node in the standing wave.

In conclusion, we have demonstrated that conical intersections may be created as a function of laboratory position space by combining an optical field and a static inhomogeneous magnetic field. This may be achieved for pairs of states of either the same or different parity, which are shifted into near-resonance by the optical field and then tuned across one another with the magnetic field. Such a CI can be created for atoms such as ^{87}Rb using realistically achievable microwave fields. The resulting Berry phase produces antiperiodic boundary conditions for states that encircle the CI, and may result in flowing states with half-integer angular momentum. At higher frequencies, optical fields might be used to produce arrays of CIs, each confined in a microtrap and interacting with one another.

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