Zeros of Rydberg–Rydberg Föster interactions

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Received 30 June 2004
Published 5 January 2005
Online at stacks.iop.org/JPhysB/38/S309

Abstract

Rydberg states of atoms are of great current interest for quantum manipulation of mesoscopic samples of atoms. Long-range Rydberg–Rydberg interactions can inhibit multiple excitations of atoms under the appropriate conditions. These interactions are strongest when resonant collisional processes give rise to long-range $C_3/R^3$ interactions. We show in this paper that even under resonant conditions $C_3$ often vanishes so that care is required to realize full dipole blockade in micron-sized atom samples.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Rydberg–Rydberg interactions are very interesting for use in mesoscopic quantum manipulations. The extremely strong interactions between two Rydberg atoms have been proposed to entangle large numbers of atoms in a non-trivial manner using the phenomenon of blockade [1]. When a mesoscopic sample is illuminated with narrowband lasers tuned to a Rydberg state, only one atom at a time can be excited if the Rydberg–Rydberg interaction exceeds the linewidth of the laser–atom coupling. Therefore, the mesoscopic cloud of atoms behaves as an effective 2-level system, with the upper level being a single collective excitation.

In order to attain the strongest possible Rydberg blockade, it is desirable to operate under conditions where the Rydberg–Rydberg interaction is not the usual $C_3/R^3$ or $C_6/R^6$ van-der-Waals interactions, but instead is resonantly enhanced by ‘Föster’ processes [1, 2] such as

$$ns + ns \rightarrow np + (n - 1)p$$

(1)

that lead to isotropic $C_3/R^3$ long-range behaviour when the $ns + ns$ states are degenerate with the $np + (n - 1)p$ states. Dramatic enhancements of collisional interactions due to such resonant couplings have been demonstrated for Rydberg excitation in a magneto-optical trap [3]. The quantum nature of these types of interactions was recently used to resolve
molecules 12 nm apart in an organic solid [4]. Due to the high density of Rydberg states, there are typically many candidate levels for such Föster processes.

An important consideration for quantum information applications of Rydberg blockade is the size of cloud required. The spatial extent of the cloud must be small enough for excitation of a single atom anywhere in the cloud to block the excitation of every other atom in the cloud. Even with the great strength of Rydberg–Rydberg interactions, the required size of the clouds is likely to be less than 10 μm for fast (1 MHz) operations. Only recently have atom clouds of such small size been produced in the laboratory [5, 6]. Even in the case that sub-micron mesoscopic samples are realized experimentally, there are other applications of such samples that benefit from the sample being as large as possible. For example, we have recently proposed single-photon sources, single-photon detectors and quantum state transmission [7, 8] using mesoscopic Rydberg blockade. In these applications one would like the cloud to be many wavelengths in size in order that the diffraction limited light fields not occupy too large a solid angle. For this to be effective requires the blockade to operate over clouds of several microns in extent.

The purpose of this paper is to examine some issues that arise in the application of Föster processes to Rydberg blockade. In particular, when the quantum numbers for the states are of the form

\[ nl + nl \rightarrow n'(l - 1) + n''(l \pm 1) \]

we show that one or more of the \( nl + nl \) states have \( C_3 = 0 \). These ‘Föster zero’ states then only exhibit the usual van-der-Waals long-range behaviour and will therefore set limits on the attainable cloud sizes for quantum manipulations using Rydberg blockade. Only when \( l' = l'' = l + 1 \) are there no Föster zero states. Recent experiments [9, 10] have observed strong suppression of excitation in very large clouds that is strong evidence for blockade but do not address whether or not the blockade is complete as required for quantum information applications.

In the following, we first (section 2) present a more detailed background discussion of the importance of strong isotropic Rydberg–Rydberg interactions for quantum manipulations using Rydberg blockade. We then illustrate in section 3 how the Föster process accomplishes this. Section 4 presents the main result of this paper: for many possible Föster processes there exists a linear combination of atomic sublevels that have \( C_3 = 0 \). This result is extended to the important case of fine-structure interactions in section 5. We conclude with more implications of this work.

2. Background

The basic physics behind the Rydberg-blockade concept is illustrated in figure 1. Suppose we have two atoms in an atomic ground state \( a \). We envision exciting the atoms to a Rydberg state \( r \) of energy \( E_r \), using a narrowband laser. As shown in the figure, excitation of one of the two atoms to the Rydberg state is allowed while excitation of the second atom is off-resonant and therefore suppressed. Addition of more atoms changes the effectiveness of this ‘blockade’ but does not change the basic physics of the situation. Excitation of more than one atom is energetically suppressed as long as the interaction between each pair of atoms in the ensemble exceeds the bandwidth of the laser–atom excitation. Neglecting spontaneous emission or decohering effects, when subject to blockade and a continuous light field the atoms will undergo coherent Rabi oscillations between states \( a + a \) and \( a + r \).

To see how this process can be used to generate interesting entanglement, we simply imagine driving the atom pair in the \(|aa\rangle\) state with a \( \pi \) pulse. The atoms will then be in a
coherent superposition of states $a$ and $r$

$$\psi(\pi) = \frac{-i}{\sqrt{2}} (|ar\rangle + |ra\rangle)$$  \hspace{1cm} (3)

that cannot be written as a product of two individual wavefunctions and is therefore entangled.

To avoid the inevitable decoherence of the unstable Rydberg state, the entanglement can be usefully transferred to a second ground state $b$ with a $\pi$-pulse from a second laser tuned to the $r \rightarrow b$ transition. Again, this concept can be extended to a collection of $N$ atoms without loss of generality. The final wavefunction is a symmetric entangled superposition of $N-1$ atoms in state $a$ and one atom in state $b$.

There are several intriguing characteristics of this process. First, the entanglement is generated between the internal states of the atom; any motion of the atom is unimportant. This means that it is not necessary for the atoms to be localized in the ground state of a confining potential, so the method does not require coherence of the external degrees of freedom of the atoms. There is also no motional constraint on the speed at which the process can occur. Second, the value of the Rydberg–Rydberg interaction $D(R)$ is not important to first order, as long as it is much larger than the bandwidth $\gamma$ of the light-atom interaction. This implies that the atoms can be at random distances $R$ from each other, as long as $D(R) \gg \gamma$. Finally, since the blockade mechanism suppresses excitation of multiple Rydberg atoms, the atoms never actually experience the strong Rydberg–Rydberg forces, avoiding heating.

Key to the entanglement process is the requirement that the Rydberg–Rydberg frequency shift be large compared to the bandwidth of the light-atom interaction. If this is violated, the fidelity of the entanglement operations will be compromised. In a mesoscopic sample, insufficient blockade in any possible excitation channel is sufficient to cause production of multiple excited atoms. We now examine the Rydberg–Rydberg interactions to see under what conditions this will be problematic.

3. Förster process

We wish to consider the interactions between two like Rydberg atoms that lead to blockade. Normally, in the absence of an external electric field these interactions are of the van-der-Waals forms $1/R^5$ or $1/R^6$. Since an electric field mixes states of opposite parity together, a Rydberg atom can have a permanent dipole moment $\langle \mathbf{p} \rangle = -e\langle r \rangle$ of magnitude as large as $-n^2ea_0$, where $e$ is the electronic charge and $a_0$ the Bohr radius, leading to a much stronger and longer range $1/R^3$ interaction. This is illustrated in figure 2. Focusing on the 50d state, for fields above 5 V cm$^{-1}$ the energy becomes linear in the field. The slope of the energy versus field
gives the dipole moment, 3300 $ea_0$ in this case. The interaction between two such atoms A and B is the familiar dipole–dipole form

$$V_{DD} = \frac{1}{R^3} (3 \mathbf{p}_A \cdot \hat{\mathbf{R}} \mathbf{p}_B - \mathbf{p}_A \cdot \mathbf{p}_B) = \frac{p^2}{R^3} (3 \cos^2 \theta - 1) \quad (4)$$

where $\theta$ is the angle between the interatomic separation $\mathbf{R}$ and the electric field $F \hat{\mathbf{z}}$. The fact that $p \sim n^2 ea_0$ makes this interaction huge for two Rydberg states. However, because the dipoles are oriented in space by the applied field, the interaction vanishes at $\theta = \cos^{-1} \sqrt{1/3} = 54.7^\circ$. This is undesirable for dipole blockade because it allows for excitation of Rydberg atom pairs located at this angle with respect to each other.

Even in the absence of a mixing electric field, it is possible to have an isotropic Rydberg atom–atom interaction of comparable strength to $V_{DD}$ if there is a degeneracy in the energy-level spectrum for a pair of atoms. For example, inspection of figure 3 shows that the 50s state of rubidium (Rb) is nearly symmetrically placed between the 49p and 50p states. This means that the 50s+50s state of a Rydberg atom pair is nearly degenerate with the 49p + 50p state. Neglecting the influence of other nearby states, the eigenstates of the two-atom system are linear combinations of 50s+50s and 49p+50p, with energy shifts proportional to $1/R^3$. Using the methods described below, we find that the Rydberg–Rydberg potential energy curves are given by

$$U_\pm(R) = \frac{\delta}{2} \pm \sqrt{\frac{4U_3(R)^2}{3} + \frac{\delta^2}{4}} \quad (5)$$

where $U_3(R) = e^2 \langle 50s||r||50p\rangle \langle 50s||r||49p\rangle / R^3 = 5.75 \times 10^3 \text{ MHz} \mu\text{m}^2 / R^3$ and the ss–pp energy defect is $\delta = E(49p) + E(50p) - 2E(50s) = -3000 \text{ MHz}$. Estimates similar to this have been previously presented by Protsenko et al [11].

A plot of $U_+(R)$ is shown in figure 4. It is isotropic (independent of $\theta$), lacking the undesirable zeros of $V_{DD}$ equation (4). If there were perfect Föster degeneracy ($\delta = 0$) $U_+(R)$
Figure 3. Energy levels for Rb near \( n = 50 \). The fine-structure splitting for the p-states is just discernable.

Figure 4. Isotropic dipole–dipole interaction for excitation to the Rb 50s state. The energy defect between the 50s+50s and 49p+50p states significantly reduces the interaction as compared to the ideal degenerate case. Going to larger \( n \) partially compensates for the non-zero energy defect.

would be of similar strength and range to \( V_{DD} \). In the example given, \( U_4(R) \) exceeds 10 MHz out to distances of 10 \( \mu m \). This would be very promising for realizing dipole blockade in quite large, optically resolvable clouds. However, as illustrated in figure 4, the energy defect for real Rb atoms reduces the interaction strength substantially, becoming the usual van-der-Waals form \(-4U_3(R)^2/3\delta\) at large distances.

An interesting possibility is to tune the energy defect to zero with an external electric field. In some cases it might be possible to do this using electric fields which are sufficiently weak that the s–p mixing would be small. However, it turns out that for Rb the quantum defects of the s- and p-states are such that the application of a small electric field increases the magnitude of \( \delta \), thus further reducing the Rydberg–Rydberg interaction strength. This is shown (for the 52s state) in figure 2.
The strength of the Rydberg–Rydberg interactions varies strongly with principal quantum number. The radial matrix elements scale as \( n^2 \), so \( U_L \) scales as \( n^6 \). Since the energy defect \( \delta \) scales as \( n^{-3} \), the van-der-Waals interactions scale as \( U_L^2 / \delta \sim n^{11/6} \). Thus the range of the Rydberg blockade increases as roughly \( n^{11/6} \), so that \( n = 70 \) has nearly twice the range as \( n = 50 \), as illustrated in figure 4. Increasing \( n \) comes with increased sensitivity to stray electric fields, black-body radiation and so forth. In practice, states up to \( n \sim 100 \) should be stable enough to perform fast quantum operations [12]. However, it would be preferable to find states with smaller energy defects and therefore larger range at smaller \( n \).

Given the lack of perfect Förster resonances for the \( s \)-state, one should consider the possibility of Förster processes in other angular momentum states where the energy defects may be smaller (or more tunable) than they are for the \( s \)-states. This topic will be taken up in the following section.

Before proceeding, we should note that we have restricted our calculations to states that are nearly resonant. Other states, such as \( 51p + 48p \) in the above example, have larger energy defects (\( \delta = 12 \) GHz for \( 51p + 48p \)) and often much smaller matrix elements as well. Adding up many such states could (and should) be done in second-order perturbation theory and will alter the \( 1/R^6 \) part of the potential somewhat but not change the overall conclusions. For the rest of this paper we will continue to neglect these other states. We will also neglect the \( 1/R^3 \) quadrupole–quadrupole interactions, and assume no electric field mixing.

4. Förster zeros

Interesting new properties of the Förster process arise when we consider higher angular momentum states. For example, consider again the \( s \)- and \( p \)-states of Rb, but this time with laser excitation of the \( p \)-states, so that the relevant Förster degeneracy is, say, \( 50p + 50p \rightarrow 50s + 51s \) with an energy defect \( \delta = 930 \) MHz. Suppose also that the atoms are subject to linearly polarized light polarized along the laboratory \( z \)-axis. We shall show that there is no Förster-induced blockade for this situation. For simplicity, let us assume that two atoms \( A \) and \( B \) are aligned with \( R = \hat{z} \), with their states denoted \( |(nlm)_A(nlm)_B \rangle, m \) being the magnetic quantum number. In this case, consider the wavefunction

\[
|\psi_0\rangle = \frac{1}{\sqrt{107}} |50p1 50p\bar{1}\rangle - \frac{1}{\sqrt{5}} |50p0 50p0\rangle + \frac{1}{\sqrt{98}} |50p\bar{1} 50p1\rangle
\]

(6)

(\( \bar{1} = -1 \)) whose matrix element of \( V_{DD} \) is zero with the \( s + s \) states:

\[
\langle 50s 51s | V_{DD} | \psi_0 \rangle = 0
\]

(7)

and so the only long-range interaction will be a van-der-Waals interaction with the comparatively far-off-resonant \( d+d \) and \( d+s \) states. Note that \( |\psi_0\rangle \) is strongly coupled to the light through its \( |50p0 50p0\rangle \) part. With strong light coupling and weak dipole–dipole coupling, we conclude that the Rb \( p \)-states will not experience long-range dipole blockade. These conclusions are not changed when \( R \) is rotated away from \( \hat{z} \). If one takes the quantization axis along \( R \) then \( |\psi_0\rangle \) stays of the same form, but each of the three parts of \( |\psi_0\rangle \) will contribute to the light-atom interaction. For the rest of the paper we shall take the quantization axis for the atomic basis states to be along \( \hat{R} \).

Another very interesting possibility from figure 3 is the nearly degenerate combination \( 48d + 48d \rightarrow 50p + 46f \) with an energy defect of only 110 MHz (neglecting fine structure for now). This has the potential for much stronger Förster interactions at large distance as compared to the \( s+s \) states. In this case there is also a wavefunction with zero coupling via \( V_{DD} \) to the \( p+f \) manifold:

\[
|\psi_0\rangle = \frac{1}{\sqrt{107}} |48d0 48d0\rangle + \sqrt{\frac{8}{107}} |48d1 48d\bar{1}\rangle + \sqrt{\frac{98}{107}} |48d2 48d2\rangle
\]

(8)
where interchange-symmetric kets are defined in terms of the quantum numbers $A = n_A l_A m_A$ of the individual atoms as $|AB⟩ = (|AB⟩ + |BA⟩)/\sqrt{2 + 2δ_{p+p}}$, and $m = -m$. We shall label such states as Förster zero states.

Whereas the Förster zero state of equation (6) can be deduced in a straightforward way essentially by inspection once the matrix elements of $s+s$ with the three $p+p$ combinations are calculated, the $d+d$ Förster zero state of equation (8) is more subtle since its matrix elements with each of the three $|pm f \bar{m}⟩$ states must vanish. Thus we now discuss in more generality the conditions for Förster zero states to exist for the process

$$nl_0 + nl_0 \rightarrow n'l_1 + n''l_2.$$  

(9)

We will assume without loss of generality that $l_1 \leq l_2$.

The Förster zero state, if it exists, is written as a linear combination $|ψ_0⟩ = \sum_{m=0}^{l_0} c(m_0) |l_0m_0 l_0\bar{m}_0⟩$, so the condition $V_{DD}|ψ_0⟩ = 0$ gives

$$\sum_{m=0}^{l_0} c(m_0) |l_1 m l_2 \bar{m}| V_{DD}|l_0m_0 l_0\bar{m}_0⟩ = 0$$  

(10)

which is effectively a generalization of equation (7) to the case where there are multiple possible final states. There are three cases of interest. For $l_1 = l_0 - 1$ and $l_2 = l_0 + 1$ (of which $d+d \rightarrow p+p$ is an example) there are $2l_1 + 1 = 2l_0 - 1$ equations in the $l_0 + 1$ unknowns $c(m_0)$. But the reflection symmetry $|l_0m_0 l_0\bar{m}_0⟩ V_{DD} |l_1 m l_2 \bar{m}⟩ = |l_0m_0 l_0\bar{m}_0⟩ V_{DD} |l_1 m l_2 \bar{m}⟩$ means that $2l_1$ of the equations are the same, leaving $l_0$ equations in $l_0 + 1$ unknowns and therefore a solution exists. For the case $l_1 = l_2 = l_0 - 1$ the same argument holds. The final case, with $l_1 = l_2 = l_0 + 1$, has equation (10) with $l_0 + 2$ equations in $l_0 + 1$ unknowns and therefore no Förster zero state.

These results can also be understood from the point of view of the molecular symmetries of the problem. Following the analysis of Marinescu [13], the Förster zero states have the molecular symmetries $3Σ_u^+$ and $1Σ_g^+$ depending on their triplet or singlet spin character. For $l_1 \neq l_2$, the orbital exchange-symmetric kets as defined above are not eigenstates of the operator $σ$, that reflects the wavefunction about a plane containing the interatomic axis. Taking into account this symmetry, we define kets

$$|l_1 m l_2 \bar{m}⟩ = |l_1 m l_2 \bar{m}⟩ + β |l_2 m l_1 \bar{m}⟩ + λ |l_1 m l_2 \bar{m}⟩ + λ β |l_2 m l_1 \bar{m}⟩$$  

(11)

which have molecular parity $(g, u)$ if $p = (1)^{l_1 + l_2 + \bar{m}} = (1, 1)$ and reflection symmetry eigenvalues $σ = β λ$, giving rise to states of traditional molecular designation $3Σ_u^+$ or $1Σ_g^+$.

Considering triplet states, and assuming $l_1 < l_2$, there are $l_1 + 1λ = β = 1 3Σ_u^+$ states, $l_1 + 1λ = 1, β = 1 3Σ_u^+$ states, $l_1 + 1λ = 1, β = 1 3Σ_u^+$ states, and $l_1 + 1λ = 1 3Σ_u^+$ states. Thus for the $d+d \rightarrow p+p$ problem, there are three $3Σ_u^+$ states with $d+d$ character but only two of $p+p$ character. Therefore, it is always possible to find a linear combination of the three $3Σ_u^+$ states that has zero coupling to the two $p+p 3Σ_u^+$ states. The reasoning is identical for the $1Σ_g^+$ symmetry.

To summarize, the isotropic $C_3/R^3$ dipole–dipole interaction generated by the Förster process $l_0 + l_0 \rightarrow l_1 + l_2$ will have states with $C_3 = 0$ unless $l_1 = l_2 = l_0 + 1$.

The above analysis has emphasized the $3Σ_u^+$ and $1Σ_g^+$ states that are degenerate in the absence of overlap. Similar reasoning shows that for the case $l_1 = l_0 - 1, l_2 = l_0 + 1$ the $3Σ_u^+$ and $1Σ_g^+$ states have no Förster zeros, nor do states of $\Lambda > 0$. In general, all of these states are coupled to the light field, but the $3Σ_u^+$ and $1Σ_g^+$ states destroy the complete blockade.
5. Fine-structure effects

In general, the fine-structure interaction cannot be neglected. At \( n = 50 \) the Rb p-state fine-structure splitting is about 800 MHz and the d splitting is 100 MHz, so that at micron-scale distances there will be strong fine-structure mixing by the dipole–dipole interaction. At long enough range, where the dipole–dipole interaction is smaller than the fine-structure splitting, we can use the same type of arguments as above to analyse the problem.

Let us consider the Föster process

\[
l_{0,j_0} + l_{0,j_0} \rightarrow l_{1,j_1} + l_{2,j_2}. \tag{12}
\]

We are mostly interested in states with total \( m_j = 0 \). As before, the \( l_{0,j_0} + l_{0,j_0} \) states are symmetric linear combinations \( |l_{0,j_0}l_{0,j_0}\tilde{m}\rangle \). For alkali atoms with half-integer \( j \) there are \( j + 1/2 \) such states. On the other hand, there are (assuming \( j_1 \leq j_2 \)) \( 2j_1 + 1|l_{1,j_1}m_0l_{2,j_2}\tilde{m}_0\rangle \) states. Half of these are removed from consideration due to the

\[
|l_{0,j_0}m_0l_{0,j_0}\tilde{m}_0\rangle V_{DD}|l_{1,j_1}m_{1}l_{2,j_2}\tilde{m}\rangle = |l_{0,j_0}m_0l_{0,j_0}\tilde{m}_0\rangle V_{DD}|l_{1,j_1}\tilde{m}l_{2,j_2}\tilde{m}\rangle
\]

symmetry. Thus the system of equations for the Föster zero amplitudes \( c(m) \) has \( j + 1/2 \) equations in \( j_1 + 1/2 \) unknowns. A normalizable solution will exist only for \( j_1 < j \). It follows that potential Föster processes such as \( nd_{3/2} + nd_{3/2} \rightarrow (n + 2)p_{3/2} + (n - 2)f_{5/2} \) will have zeros.

The inclusion of fine structure brings new possibilities, however, since states of different \( l \) can have the same value of \( j \). For example,

\[
d_{3/2} + d_{3/2} \rightarrow (n + 2)p_{3/2} + (n - 2)f_{5/2} \tag{13}
\]

has an energy defect of only \(-15\) MHz at \( n = 39 \) and should not have any Föster zeros. The Hamiltonian matrix has the structure

\[
H = \begin{pmatrix}
\text{diag}(0) & WU_3(R) \\
WU_3(R) & \text{diag}(\delta)
\end{pmatrix}
\tag{14}
\]

where the interaction submatrix

\[
W = \begin{pmatrix}
-\frac{4}{25}\sqrt{\frac{2}{3}} & 0 & 0 & \frac{4}{25}\sqrt{\frac{2}{3}} \\
\frac{8}{25}\sqrt{\frac{2}{3}} & \frac{4}{15} & \frac{-4}{25}\sqrt{\frac{2}{3}} & 0 \\
\frac{8}{25}\sqrt{\frac{2}{3}} & \frac{-8}{25}\sqrt{\frac{2}{3}} & \frac{-4}{15} & 0 \\
\end{pmatrix}
\tag{15}
\]
and \( U_3(R) = e^2 \langle 39d||r||41p\rangle \langle 39d||r||37f\rangle / R^3 = 2940 \text{ MHz } \mu \text{m}^3 / R^3 \). For \( \delta = 0 \), the eigenvalues of \( \mathbf{H} \) corresponding to the d+d states are \((\pm 4 \sqrt{31} \pm \sqrt{937} / 75) U_3(R)\). Two of the eigenvalues are quite small \((\pm 0.033 U_3(R))\), leading to poor Rydberg blockade. The potential curves for \( \delta = -15 \text{ MHz} \) are shown in figure 5. The nearly flat potential shows that while there are no F"oser zeros for this case, the Rydberg blockade is still strongly suppressed at large distances even though the energy defect is very small. Even in the presence of fine structure, the blockade is still poor if \( l_1 < l_0 \).

6. Conclusions

The primary result of this paper is that the very long-range \( C_3 / R^3 \) interactions produced by resonances \( n l + n l \rightarrow n' l_1 + n'' l_2 \) have states with \( C_3 = 0 \) unless \( l_1 = l_2 = l + 1 \). This strongly reduces the number of possibilities for attaining high fidelity dipole-blockade in mesoscopic atom samples. One solution is to rely instead on quadrupole–quadrupole \((1 / R^5)\) or second-order dipole–dipole \((1 / R^6)\) interactions to achieve blockade at high values of \( n \). As pointed out recently by the Connecticut group [14, 9] the interactions under these conditions can be quite strong.

Another possibility for attaining strong dipole blockade is to tune \( l_1 = l_2 = l + 1 \) resonances with an electric field. For example, the great sensitivity of f states to electric fields can tune d+d→f+f into resonance at modest fields. At resonance, the long-range potentials for this case are \( \pm 0.336 U_3, \pm 0.227 U_3, \pm 0.158 U_3 \) and thus should lead to quite strong Rydberg blockade at long range.

An important topic for further study is the sensitivity of the F"oser zeros to electric fields. If the Stark splitting of the p or d magnetic sublevels is greater than \( U_3 \), we would expect the zeros to no longer be present. However, an important question is whether the Rydberg–Rydberg interaction becomes anisotropic in this case, since the atoms would also acquire a permanent, spatially oriented dipole moment.

Acknowledgments

The authors recognize very helpful conversations with other members of the Wisconsin AMO Physics group, and with C Goebel and R Cote. This work was supported by the National Science Foundation, NASA, and the Army Research Office.

Appendix

A.1 Calculation of dipole–dipole matrix elements

Let \( l_1, l_2, l \geq l_1 \) be the angular momenta involved in the F"oser process

\[
\frac{nl + nl}{nl + nl} \rightarrow n' l_1 + n'' l_2. \tag{A.1}
\]

Since the dipole–dipole interaction

\[
V_{DD} = \frac{\sqrt{6} e^2}{R^3} \sum_p c_{np}^{(2)} r_{Ap} r_{Bp}, \tag{A.2}
\]

expressed in a coordinate system with \( z \) aligned with \( \mathbf{R} \), is symmetric on atom interchange \((A \leftrightarrow B)\), the interchange-symmetric state

\[
|lm\tilde{m}\rangle \equiv \frac{|lm\rangle_A (lm')_B + |lm'angle_A (lm)_B}{\sqrt{2(1 + \delta_{mm'})}} \tag{A.3}
\]
mixes only with the symmetric combinations
\[ |l_0m_0 l_2m_2⟩ = |l_0m_0 l_2m_2 + l_2m_2 l_0m_0⟩/\sqrt{2}. \tag{A.4} \]
The matrix element of \( V_{DD} \) is therefore
\[ \langle lm'm' | V_{DD} | l,m l_2m_2⟩ = \langle lm'm' | V_{DD} | l,m l_2m_2⟩ + (m \leftrightarrow m')/\sqrt{1 + \delta_{mm'}}. \tag{A.5} \]
We use the Wigner–Eckart theorem to write this explicitly as
\[ \langle lm'm' | V_{DD} | l,m l_2m_2⟩ = U_3(R) \sum_p C_{1p1p}^{l,m} C_{2p1p}^{lm} + (2 \leftrightarrow 1)/6 \tag{A.6} \]
where
\[ U_3(R) = \frac{e^2 |nl⟩⟨n'l'_l|}{R^3} \tag{A.7} \]
and the reduced matrix elements are given in terms of radial integrals of \( r \) as
\[ \langle n|l⟩⟨n|l'⟩ = \sqrt{2l + 1} C_{i00}^{l} \int_0^∞ r P_n(r) P_{l,l'}(r)dr. \tag{A.8} \]
For the calculation of matrix elements in this paper, we have used the \( l \)-dependent core potentials of Marinescu \textit{et al} [15] and obtained the radial wavefunctions by Numerov integration of the Schrödinger equation. Energy levels were calculated using the recent quantum defect determinations of Li \textit{et al} [16].

A.2. Reflection symmetry
The dipole–dipole matrix element \( \langle l_0m_0 l_0m_0 | V_{DD} | l,m l_2m_2⟩ \) is proportional to
\[ \sum_p \left( C_{1m1p}^{l_0m_0} C_{1p1p}^{20} C_{2p1p}^{l_0m_0} + C_{2m1p}^{l_0m_0} C_{1p1p}^{20} C_{2p1p}^{l_0m_0} \right) \]
\[ = \sum_p \left( C_{1m1p}^{l_0m_0} C_{1p1p}^{20} C_{2p1p}^{l_0m_0} + C_{2m1p}^{l_0m_0} C_{1p1p}^{20} C_{2p1p}^{l_0m_0} \right) \tag{A.9} \]
where the Clebsch–Gordan symmetry \( C_{a+b}^{γγ} = -1^{a+b−c} C_{a+b}^{γγ} \) has been used along with the specific parities \((-1)^l = (-1)^l = -(-1)^l \). Inspection of equation (A.9) then shows that
\[ \langle l_0m_0 l_0m_0 | V_{DD} | l,m l_2m_2⟩ = \langle l_0m_0 l_0m_0 | V_{DD} | l,m l_2m_2⟩. \tag{A.10} \]

References