Alignment of s-state Rydberg molecules in magnetic fields

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We unravel some peculiar properties of ultralong-range Rydberg molecules formed by an s-state ⁸⁷Rb Rydberg atom and a corresponding ground-state atom whose electronic orbitals are spherically symmetric and therefore should not be influenced by the presence of weak magnetic fields. However, the electron-atom interaction, which establishes the molecular bond, is under certain conditions subject to a sizeable spin-orbit coupling and, hence, sensitive to the magnetic field. This mechanism can be harnessed to counterintuitively align the s-state molecules with respect to the field axis. We demonstrate this by analyzing the angular-dependent Born-Oppenheimer potential energy surfaces and the supported vibrational molecular states. Our predictions open novel possibilities to access the physics of relativistic electron-atom scattering experimentally.

Introduction - The extraordinary properties of ultralong-range Rydberg molecules (ULRM) such as their enormous size and large permanent electric dipole moments (PEDM) have stirred increasing interest in their study. Two main molecular species can be distinguished. Nonpolar molecules with low electronic angular momentum, e.g. s-state or d-state ULRM, and polar ones with high angular momentum. The latter are called Trilobite and Butterfly molecules after their electronic wave function shaped by s- and p-wave interactions [1, 2].

In recent years a multitude of spectroscopic experiments have probed ULRM from their first detection in Rubidium [3], Cesium [4], and Strontium [5], their coherent excitation [6], lifetime measurements [7–9], the observation of PEDM [10, 11] and angular momentum couplings [12], to proving the existence of polymers [13, 14] and controlling ULRM in external fields [15–17]. Additionally, several applications have been proposed and implemented such as probing lattice gases via ULRM [18] and performing remote spin flips [19], as well as the study of ultracold chemical reactions [20] and electromagnetically induced transparency [21]. Moreover, they provide the possibility to realize Rydberg and ionic impurities in Bose-Einstein condensates [22–27], and to study ion-atom scattering [28] as well as to employ optical Feshbach resonances in order to tune atom-atom interaction [29, 30].

The molecular binding originates from s- and p-wave scattering of the Rydberg electron off a neutral ground-state atom [31, 32]. Therefore, ULRM serve as a unique tool to characterize electronic scattering channels [33, 34] and resonances at very low electronic energies [35, 36]. Specifically they provide an excellent platform for experimentally testing ab initio calculations of s- and p-wave scattering phase shifts [37–40]. In particular, in Rubidium and Cesium the electron-neutral scattering possesses a p-wave shape resonance, which can be understood as a short-lived, meta-stable, anionic state of the electron-neutral system [41–43]. A higher level theory that takes relativistic spin-orbit coupling of the two valence electron’s spin with the their angular momentum relative to the ground-state atom’s core into account has been provided [44–46].

In this letter, we demonstrate that the relativistic spin-orbit interaction paired with a weak magnetic field gives rise to the surprising feature of alignment of s-state ULRM. For d-state ULRM, alignment in magnetic fields has been demonstrated and stems from the nonzero orbital angular momentum of the electronic state [15, 47]. However, for s-state, i.e. zero angular momentum, ULRM such an alignment process is unexpected, due to the isotropy of the electronic state. Indeed for s-state ULRM, the alignment originates from the interaction of the electronic spin-degrees of freedom with the spatial degrees of freedom within the p-wave scattering. A pictorial illustration of the mechanism is presented in figure 1: In a first step, the magnetic field couples to the electronic spins and tends to align them. In a second step, the spin-orbit coupling modifies the electron-atom interaction in an angular-dependent fashion.

Figure 1. Pictorial representation of the alignment mechanism of the Rydberg electron and the ground-state atom. The magnetic field (gray arrows) tends to orient the electronic spins (black arrows). Due to spin-orbit interaction, the relative angle w.r.t. the internuclear axis θ influences the scattering process (orange arrows) and leads, consequently, to angular-dependent potential energy surfaces.
Figure 2. Sketch of the molecule and its spin degrees of freedom in two different representations. In (a) the ground state atom with nuclear spin $I$ and electronic spin $\hat{s}_2$ is located outside the electronic cloud of the Rydberg atom (yellow shading). The Rydberg atom’s core is located in the center and the Rydberg electron at position $\vec{r}$ carries orbital angular momentum $\vec{I}$ and spin $\hat{s}_1$. In (b) the ground state atom is located inside the cloud at position $\vec{R}$. Here, the electronic spins are coupled to the total electronic spin $\hat{S}$ which again couples to the electron pair’s angular momentum relative to the ground-state atom’s core $\vec{L}$.

We analyze this alignment by determining the angular-dependent Born-Oppenheimer potential energy surfaces (PES) as well as energies and densities of bound vibrational states. To provide a more intuitive physical picture of the underlying mechanism, we present an approach that considers the presence of $p$-wave shape resonances and the magnetic field perturbatively and reproduces the polar potential energy curves of the underlying ab initio calculations. We focus here on Rubidium molecules. However, our approach applies also to ULRM of other atomic species that possess a $p$-wave shape resonance such as Cesium.

Setup and Interactions - Our diatomic ULRM consists of a Rydberg and a ground-state Rubidium atom. The closed shell core electrons of the Rydberg atom are taken into account by quantum defects of the Rydberg energy levels best obtained by experimental observation [48, 49]. The polarizable ground-state atom with hyperfine structure [50] at relative position $\vec{R}$ to the Rydberg core acts as a perturber to the Rydberg electron’s wave function (see figure 2 for a sketch including all relevant spin degrees of freedom). The interaction of the ground-state atom with the Rydberg electron can be described via a generalized Fermi pseudo-potential

$$
\hat{H}_B = \vec{B} \cdot \left( \hat{S} + \vec{l}/2 \right),
$$

where $l$ is the Rydberg electron’s orbital angular momentum relative to the Rydberg core, which is nonzero for states other than the $s$ state.

The electronic Hamiltonian [46, 47] is diagonalized within the Born-Oppenheimer approximation, yielding the adiabatic PES which depend parametrically on the distance between the Rydberg core and the ground-state atom. Indeed, due to the azimuthal molecular symmetry corresponding to rotations around the magnetic field axis, the PES depend only on the internuclear distance $R$ and the polar angle $\theta$ between the magnetic field axis and the internuclear axis. Our basis includes the closest hydrogenic manifold, which lies energetically above the considered $s$ state (compare figure 3), with all Rydberg electronic angular momentum states that have a maximal projection onto the internuclear axis of $m_{\text{max}} = 3/2$, and all ground-state atomic hyperfine states. Further increase of $m_{\text{max}}$ has only minor influence on our results (c.f. [51]).

Results - Figure 3 depicts a radial cut of the PES for Rubidium 28s (sPES) and $F = 2$ in a magnetic field of $B = 10$ G that is aligned parallel to the internuclear axis. The presence of the neutral ground-state atom in the Rydberg electronic cloud leads to an oscillatory potential curve typical for ULRM. Illustratively speaking, the emerging potential wells act as traps for the neutral ground-state atom and lead to radially localized bound states between the Rydberg atom and the ground-state atom, thus, forming a molecule. At distances $R \approx 800a_0$, where $a_0$ is the Bohr radius, the kinetic energy of the Rydberg electron matches the energy of the $p$-wave shape resonances, which leads to a steep crossing of the butterfly PES. Sufficiently far from the resonance, the PES can be characterized by their total electronic spin character, which is shown in figure 3 as a colorcode. Deep potential
Figure 3. Radial cut of the PES for a Rubidium 28s state in a magnetic field of $B = 10$ G parallel to the internuclear axis ($\theta = 0$) for the ground-state atom in a hyperfine $F = 2$ state. The colorcode indicates the expected total electronic spin. A pure triplet curve with 6-fold multiplicity is visible as well as a mixed singlet-triplet curve 4-fold multiplicity (inset). In the deepest potential well around $R = 870$ a$_0$ close to the p-wave shape resonance (see encircled region) the Zeeman splitting is not equidistant.

wells occur, when the electronic spins are aligned to a triplet state ($S = 1$), and shallow wells, when the spins are anti-aligned in a mixed singlet-triplet state. The presence of the magnetic field splits the otherwise degenerate curves and reveals their multiplicity, which in the case of the $F = 2$ hyperfine state is six for the triplet curve and four for the mixed curve. Typically within a region of 100 a$_0$ around the crossing with the butterfly curve, we find a significant dependence of the sPES on the polar angle $\theta$. An angular cut of the sPES at the radial position of the deepest potential well at $R = 870$ a$_0$ is shown in gray in figure 4(a). The emerging angular potential wells are deep enough to support aligned molecular states for which three equilibrium angles are possible. Both the energetically lowest and highest visible sPES allow for molecules aligned perpendicular to the magnetic field axis ($\theta = \frac{\pi}{2}$), whereas all other visible sPES allow for parallel and anti-parallel alignment ($\theta = \{0, \pi\}$). The second lowest visible sPES shown in gray in figure 4(a) constitutes an exception to this and is mostly flat not allowing any alignment. The angular dependence of the sPES is only present, if the spin-orbit interaction is included in the description and the p-wave interaction is sufficiently strong. When choosing equal scattering lengths for all three triplet p-wave channels and therefore effectively eliminating the relativistic spin-orbit coupling in the scattering event, the polar-angle dependence vanishes, which is shown in light blue. Stationary solutions of the vibrational Schrödinger equation with the Hamiltonian

$$\hat{H}_{\text{vib}} = -\frac{1}{M}(\partial_{\rho}^2 + \partial_z^2) + \frac{m^2 - 1/4}{M\rho^2} + \epsilon_i(\rho, z),$$

Figure 4. (a) Angular cut of the PES for a Rubidium 28s state in a magnetic field of $B = 10$ G at the radial position $R = 870$ a$_0$ matching the position of the potential well close to the p-wave shape resonance (compare figure 3). The numerical result within exact diagonalization of the electronic Hamiltonian (light blue and gray lines) is compared to a reduced model only taking the six relevant states of the $K = 5/2$ multiplet into account (blue and black dashed lines). Both approaches are considered with equal (light blue, blue) and distinguishable (gray, black) p-wave scattering lengths. (b)-(c) Density plots in cylindrical coordinates of vibrational states supported by the $m_F = -2$ and $m_F = 0$-dominated state, respectively. The ground-state atom localizes in the perpendicular (parallel) configuration relative to the magnetic field axis. The eigenstate energy $\epsilon_\nu$ is provided relative to the unperturbed 28s, $F = 2$ atomic energy. The vibrational ground state features an alignment of $\langle \cos^2 \theta \rangle = 0.04$ (b) and 0.98 (c).
are obtained via a two-dimensional finite difference method with the rotational angular momenta around the internuclear axis m set to zero [15, 47]. Here, M is the atomic mass of $^{87}$Rb and $c_i(ρ, z)$ are the PES. For the energetically lowest curve in figure 4(a), which represents the spin polarized $F = 2, m_F = -2$ state, the densities of the vibrational ground state and the first three excited states are provided in cylindrical coordinates in figure 4(b). They exhibit an energy spacing of 1 MHz and the vibrational ground state features an alignment of $⟨\cos^2 θ⟩ = 0.04$. The third gray curve from the bottom in figure 4(a) corresponds to an s state dominated by $F = 2, m_F = 0$ contributions and possesses potential wells around $θ = 0$ and $θ = π$. The densities of the corresponding vibrational states can be seen in figure 4(c) and exhibit an energy spacing of 2 MHz, while the vibrational ground state features an alignment of $⟨\cos^2 θ⟩ = 0.98$.

Discussion - In order to gain insight into the effect of the spin-orbit interaction, we develop a perturbative approach. To reproduce the angular cuts of the PES, it is sufficient to restrict the Hilbert space to a subspace with constant quantum number $K = |\vec{S} + \vec{I}|$, which represents the molecular system’s total angular momentum. The extreme values for $K = \{\frac{1}{2}, \frac{5}{2}\}$ correspond to pure triplet ($S = 1$) states, whereas $K = \frac{3}{2}$ states have mixed singlet and triplet character. The Hamiltonian then reads

$$\hat{H}(θ) = c_s + c_p \sum_{J=0}^2 \hat{V}_p(J) + \hat{H}_B,$$

where $c_s$ and $c_p$ are parameters which are adjusted to reproduce the results of the field-free exact diagonalization. $c_s$ corresponds to the overall offset due to the dominant s-wave interaction and $c_p$ controls the admixture of the p-wave interaction. The p-wave interaction depends on the total electronic angular momentum relative to the ground-state atom $J$ and reads

$$\hat{V}_p(J) = a(\rho, J, K) \sum_{M_p} |C^{JM_p}_{0,0,130}M_p|^2 |M_S⟩⟨M_S|,$$

where $M_S$ are the three possible projections of $S = 1$ and $C^{JM_p}_{LM_p,SM_p}$ is a Clebsch-Gordan coefficient describing the coupling of $\vec{S}$ and $\vec{L}$ to $\vec{J}$. The interaction matrix is diagonal for each $J$-dependent channel. Equation (5) was derived using simplifications valid for Rydberg s-states, however, it can be generalized to describe also other states with non-zero angular momentum.

The presence of a magnetic field acting on the electronic state of zero orbital angular momentum $l = 0$ with rotational freedom, w.l.o.g. around the y axis, is reflected by

$$\hat{H}_B = B \left(\hat{S}_z \cos θ + \hat{S}_x \sin θ\right),$$

with Zeeman splitting proportional to the magnetic field $B$ and the spin matrices for $z$ and $x$ direction $\hat{S}_{z,x}$. If the scattering lengths for each p-wave channel are equal, the generator of rotations around the y axis $\hat{S}_y$ commutes with $\hat{V}_p$, therefore the eigenvalues of $\hat{H}(θ)$ are independent of $θ$. By choosing different p-wave triplet scattering lengths, the spin-orbit interaction can effectively be switched on and off. Solving $⟨KM_K|\hat{H}(θ)|KM_{K′}⟩$, where $M_K$ is the projection of $K$ onto the internuclear axis, reproduces the correct number of PES for s-state molecules and reduces the problem to diagonalizing a $(2K + 1)$-square matrix, whereas the original diagonalization including the neighboring hydrogenic manifold involves 1488 basis states.

Figure 4(a) shows the sPES of the model in the case of $K = 5/2$ for a magnetic field of $B = 10$ G, which corresponds the curves circled in red in figure 3. For distinguishable (equal) scattering lengths, shown in black (blue) the model recovers the results of the exact diagonalization, which are shown in grey (light blue).

For larger magnetic fields, when $\hat{H}(θ)$ is dominated by $\hat{H}_B$, the different PES separate further while the depth of the potential wells does not increase. For smaller magnetic fields, when $\hat{H}(θ)$ is dominated by $\hat{V}_p(J)$, the PES are structured in pairs of equal absolute total angular momentum projection $|M_K|$. The $θ$-dependent influence of the interaction matrices $\hat{V}_p(J)$, which represent the three different triplet p-wave scattering channels, on the eigenvalues of $\hat{H}(θ)$ corresponds to the physical picture that the spin-orbit interaction introduces a spatial degree of freedom and consequently angular anisotropy to the otherwise isotropic molecular system.

Conclusion - We predict the possibility of molecular alignment of s-state ultralong-range Rydberg molecules in a homogeneous magnetic field. In contrast to d-state ULRM, that can be aligned in magnetic fields due to the nonzero orbital angular momentum of the Rydberg state, the alignment of s-state ULRM has a completely different origin. We attribute the effect to the interplay of a magnetic field and the spin-orbit coupling of the electron-atom interaction. For the alignment to occur in s-states, it needs both, the $\vec{L} \cdot \vec{S}$- and the strong p-wave interaction. The alignment mechanism can be interpreted by means of a reduced interaction model in a comparably low dimensional Hilbert space. This novel degree of control of the otherwise isotropic s-states of ULRM is clearly within reach of current experimental efforts. Beyond this work, it would be beneficial to study the influence of higher angular momentum states on the alignment, for instance in trilobites or p-state molecules, with the butterfly molecules being a prominent candidate to show effects of spin-orbit interaction. Furthermore similar effects should occur in polyatomic ULRM and are expected to lead to novel forms of angular dependent three-body interactions. ULRM are an exceptionally suitable environment to experimentally study low-energy electronic scattering. Especially, the study of spin-orbit interaction effects provides unique possibilities to exper-
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