Ultralong-range Rydberg bi-molecules

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We predict that ultralong-range Rydberg bi-molecules form in collisions between polar molecules in cold and ultracold settings. The collision of Λ-doublet nitric oxide (NO) with long-lived Rydberg NO(\(n_f, n_g\)) molecules forms ultralong-range Rydberg bi-molecules with GHz energies and kilo-Debye permanent electric dipole moments. The Hamiltonian includes both the anisotropic charge-molecular dipole interaction and the electron-NO scattering. The rotational constant for the Rydberg bi-molecules is in the MHz range, allowing for microwave spectroscopy of rotational transitions in Rydberg bi-molecules. Considerable orientation of NO dipole can be achieved. The Rydberg molecules described here hold promise for studies of a special class of long-range bi-molecular interactions.

Rydberg spectroscopy is one of the most precise techniques in the AMO physics toolkit to probe core interactions, perform cavity quantum electrodynamics [1], diagnose environmental influence in laboratory and astrophysical gases [2, 3], engineer strong quantum correlations, and transmit quantum entanglement over macroscopic distances [4]. In cold and pristine ultracold AMO traps, Rydberg interactions with surrounding atoms or molecules may, under certain conditions, lead to the formation of special long-range Rydberg molecules [5, 6]. Such Rydberg molecules have proven to be fertile grounds for investigating novel few-body phenomena, many-body quantum correlations [7–9], and exotic ionic interactions [10, 11]. To date, ultracold Rydberg excitations have been performed in a target atom- usually an alkali metal or alkaline earth atom [8, 12].

Ultracold polar molecules have accessible internal degrees of freedom, which can be engineered for simulations of chemical reactions [13–15], many-body interactions [16, 17], and information processing in the quantum limit [18, 19]. As with Rydberg systems, polar molecules can possess large dipole moments, making them amenable to control and manipulation. In addition, interactions involving molecules are fundamental to chemical synthesis, whose studies are in vogue in cold and ultracold traps as densities increase [20–23]. In most cold and ultracold settings, atom-molecule or molecule-molecule collisions often are exothermic, releasing energy leading to losses from the trap. Elaborate schemes to control and shield trapped molecules from such deleterious encounters are proposed [24–26].

Here, we investigate a particular class of bi-molecular collision, facilitated by Rydberg excitation in a Λ-doublet molecule(NO), leading to formation of exotic Rydberg bi-molecules, whose electronic and rovibrational properties can be readily controlled. Because the Λ-doublet transitions can be precisely measured, such molecules are leading candidates for searches for the variation of fine-structure constant and electron to proton mass ratio [27].

Nitric oxide is a Λ-doublet molecule, and has been extensively studied for its numerous applications across physics and chemistry. NO is an atmospheric constituent, which plays a large role in the chemistry and heat budget of the thermosphere, and at lower altitudes is a catalyst in depleting atmospheric ozone [28]. Because NO is also a neurotransmitter and neurotoxic [29, 30], there is practical interest in NO detection. A recent promising technique for detection of NO concentration with high sensitivity is through Rydberg excitation in NO [31].

There are a number of unique features of Rydberg NO-NO bi-molecular interactions at large distances, see Fig. 1: a) hybridization of nearby Λ-doublet opposite parity states is efficient even when molecules possess permanent electric dipole moments (PEDM) much below the Fermi-Teller critical dipole, \(d_c = 1.63D\) [32],...
is 236 meV \[41\].

1) resonances, which appear in the inelastic channels.

NO resonance at \(\sim\) 100 meV. This resonance has been measured \[39, 40\] in the channel \(\nu = 0\) scattering resonance. Electron scattering \(|\nu\rangle \rightarrow |\nu'\rangle\) can form negative ions, NO\(^{−}\).

The Hamiltonian \(H_{\text{e−NO}}\) contains the Rydberg electron and core interacting with the NO PEDM and the scattering of the Rydberg electron from NO,

\[
H_{\text{e−NO}}(\mathbf{r}, \mathbf{R}) = -d_{\text{NO}} \cdot \mathbf{F}_{e}(\mathbf{r}, \mathbf{R}) + 2\pi\alpha_{e}(k)\delta(\mathbf{r} - \mathbf{R}) + 6\pi\alpha_{e}^{3}(k)\delta(\mathbf{r}) \cdot \nabla \cdot \nabla,
\]

where \(\mathbf{r}\) and \(\mathbf{R}\) are the positions of the Rydberg electron and NO\((X^{2}\Pi_{1/2})\) with respect to the core NO\(^{+}\), respectively. The first term in Eq. (2) describes the interaction of the NO\(^{+}\) core and the Rydberg electron with the NO\((X^{2}\Pi_{1/2})\) PEDM \(d_{\text{NO}}\). The internal field due to the Rydberg electron and core, \(F_{e}(\mathbf{R}, \mathbf{r}) = e [\mathbf{r} - \mathbf{R}/(\mathbf{r} - \mathbf{R})] \cdot \nabla\), hybridizes the \(\Lambda\)-doublet states \(|+\rangle\) and \(|−\rangle\) of NO\((X^{2}\Pi_{1/2})\). The total angular momentum excluding spin couples to \(F_{z,e}(\mathbf{R}, \mathbf{r})\), while the other two perpendicular components of this electric field do not contribute to the interaction. We further assume that the position of NO\((X^{2}\Pi_{1/2})\) is fixed along the \(Z\)-axis.

The last two terms in Eq. (2) describe the Rydberg electron collision with NO. These two terms, respectively, reflect the contributions to the low-energy electron-molecule scattering in the \(L = 0\) (\(S\)-wave) and \(L = 1\) (\(P\)-wave) scattering partial waves \[12, 36, 37\]. The \(S\)-wave scattering length in the Fermi pseudopotential is, \(a_{S}(k) = -\tan(\delta_{S}(k))/k\), with \(\delta_{S}(k)\) the \(L = 0\) scattering phase shift, and the scattering volume, \(a_{S}^{3}(k) = -\tan(\delta_{R}(k))/k^{3}\), diverges at the position of an \(L = 1\) scattering resonance. Electron scattering from NO\((X^{2}\Pi_{1/2})\) can form negative ions, NO\(^{−}\)(\(X^{3}\Sigma^{−}\)). The electron affinity of NO has been measured by Alle et al. \[38\], \(E_{a} = 33 \pm 10\) meV. In elastic electron scattering from NO\((X^{2}\Pi_{1/2}(\nu = 0))\), there exists a low-energy resonance at \(\sim 100\) meV. This resonance has been measured \[39, 40\] in the channel \([e^{-} + \text{NO}(X^{2}\Pi_{1/2}(\nu = 0)) \rightarrow \text{NO}^{−}(X^{3}\Sigma(\nu' = 1))]\); there are additional NO\(^{−}\)(\(X^{3}\Sigma(\nu' > 1))\) resonances, which appear in the inelastic channels. The fundamental vibrational frequency in NO\((X^{2}\Pi_{1/2})\) is 236 meV \[41\].

Both NO and NO\(^{+}\) have PEDM, and the resulting dipole-dipole interaction has also been incorporated in the Hamiltonian (1). For the NO\((nl)\)-NO bi-molecules, at separations of hundreds \(a_{0}\), these dipolar interactions are several orders of magnitude smaller that the dominant charge-dipole interactions.

The wave function of Hamiltonian (1) is expanded in electronic Rydberg NO and molecular doublet basis and the corresponding Schrodinger equation, \[HC = U_{\text{bi-mol}}(R)\mathbf{C},\] is solved to obtain the bi-molecular Rydberg BO potential energy curves \(U_{\text{bi-mol}}(R)\), and the electronic wave function \(\mathbf{C}\).

To obtain the wave functions and energies for the Rydberg NO, a quantum defect description is appropriate. The Rydberg NO BO potential can be written as

\[
U_{\Lambda}(R_{\text{NO}}) = U^{+}(R_{\text{NO}}) - \frac{1}{2(n - \mu_{\Lambda})^{2}} ,
\]

where \(U^{+}(R_{\text{NO}})\) is the ground-state BO potential curve for NO\(^{+}\)(\(X^{1}\Sigma^{+}\)) and \(\Lambda\) is the projection of Rydberg electron orbital quantum number \(l\) on \(R_{\text{NO}}\). The quantum defect \(\mu_{\Lambda}\) depends on \(n\) and \(R_{\text{NO}}\). In our analysis, \(R_{\text{NO}}\) is fixed at the equilibrium separation, \(R_{\text{NO}} = 2.175\ a_{0}\). The quantum defects were calculated by Rabadán and Tennyson using a 12 state R-matrix method \[42\]. The Rydberg electron wave functions and energies for different molecular symmetries were obtained by solving the hydrogenic Schrodinger equation for non-integer values of \(n\) \[43, 45\] using the finite element method. We note that frame transformation multichannel quantum defect theory \[46\] is a compact formulation of Rydberg electron interaction with molecular core electrons. In Rydberg NO, for instance, it results in mixing of Hund’s cases (\(d\)) and (\(b\)) molecular symmetries. This mixing is particularly important at low orbital angular momenta, when the electron spends considerable time near the core and Rydberg energies will depend on the ion rotational quantum numbers \[47\]. Here, we are interested in formation of ultralong-range Rydberg bi-molecules when the NO molecules are excited into high orbital angular momentum states, \(nf\) and \(ng\).

Fig. 2 (a) presents the BO potential energy curves when only the charge-dipole interaction is included in the Hamiltonian (2) \[48, 49\]. The near-degenerate \(\Lambda\)-doublet combined with the small quantum-defects of the \(nf\) and \(ng\) Rydberg states give rise to several potential wells supporting bound vibrational levels. By taking into account the low energy scattering of the Rydberg electron with NO, the complexity of these electronic states is significantly enhanced in Fig. 2 (b). Because of the interplay of charge-dipole interaction and \(e^{-}\)-NO collision, deep molecular binding occurs at large distances (\(R \sim 500\ a_{0}\)), in the exponential tail of NO\((15f)\) and NO\((15g)\) Rydberg wave functions, see the NO\((15f)\)-NO\((X^{2}\Pi_{1/2})\) and NO\((15g)\)-NO\((X^{2}\Pi_{1/2})\) BO potential curves in Fig. 2 (c) and (d), respectively. The outer wells support bound levels with 200 – 800 MHz vibrational spacing. At smaller distances, \(R \lesssim 400\ a_{0}\), the \(P\)-wave resonance intro-
FIG. 2. BO potential energy curves of the NO($n = 15, l \geq 3$)-NO($X^2\Pi_{1/2}$) ultralong-range Rydberg bi-molecule including (a) the charge-dipole only and (b) all the interactions in the Hamiltonian (2). Details of the curves including all interactions near the (c) NO($15g$)-NO($X^2\Pi_{1/2}$) and (d) NO($15f$)-NO($X^2\Pi_{1/2}$) dissociation thresholds; the vibrational wave functions are shown at the energy level. The avoided crossing in (c) are quantum reflected states [50].

FIG. 3. (a) BO potential energy curves of the NO($n = 40, l \geq 3$)-NO($X^2\Pi_{1/2}$) Rydberg bi-molecule including all the interactions in the Hamiltonian (2). Details of the curves near the (b) NO($15g$)-NO($X^2\Pi_{1/2}$) and (c) NO($15f$)-NO($X^2\Pi_{1/2}$) dissociation thresholds; the vibrational wave functions are shown at the energy level.

... reduces avoided crossing and $l$–mixing among the potential energy curves, destroying several inner wells visible in Fig. 2 (a).

Because of proximity to hydrogenic degenerate manifolds, the $nf$ and $ng$ Rydberg states have considerable $l$-mixed character, giving rise to long lifetimes. For the $nf$ states, lifetimes of $10 - 50$ ns have been measured [51], whereas $ng$ states have lifetimes of $0.15 \mu s < \tau < 0.6 \mu s$ for $30g < nl < 55g$, against predissociation and vibrational autoionization [52]. Due to the $l$-mixing, the NO($15f$)-NO($X^2\Pi_{1/2}$) and NO($15g$)-NO($X^2\Pi_{1/2}$) bi-molecules possess fairly large PEDM, $\sim 0.5$ kD. The rotational constant for the $n = 15$ Rydberg NO bi-molecules is about $0.5$ MHz. As a consequence, microwave spectroscopy of rotational transitions now becomes feasible, and realistic microwave Rabi frequencies of $1 - 10$ MHz can be achieved.

The BO potential energy curves converging to the NO($40f$)-NO($X^2\Pi_{1/2}$) dissociation threshold, are shown in Fig. 3. Because the $P$-wave resonance affects the energies at shorter distances, its effect on the NO($40f$) and NO($40g$) Rydberg bi-molecular states is reduced. The potential curves converging to NO($40f$) in Figs. 3 (b) and (c) support deep vibrational levels. The PEDMs for NO($40f$)-NO($X^2\Pi_{1/2}$) and NO($40g$)-NO($X^2\Pi_{1/2}$) electronic states are in the $5 - 7$ kD range. The $A$-doublet separated upper curve in NO($40f$), for instance, in Fig. 3(b) has a double-well with well-separated molecular states. The right and left wells correspond to NO molecular orientation away and toward the Rydberg molecular core [48]. The degree of NO dipole orientation is roughly $40\%$. This orientation is even larger for the NO($40g$)-NO($X^2\Pi_{1/2}$) bi-molecules in Fig. 1.

The Rydberg bi-molecules can be experimentally created by excitation of the main optical transition in NO, $A^2\Sigma^+ (\nu' = 0, N' = 0) \leftarrow X^2\Pi (\nu = 0, N = 0)$ at $225$ nm in the $\gamma$-band [31, 47, 53]. The $A^2\Sigma^+$ state has configuration interaction coefficients, $94\% (l = 0)$, $1\% (l = 1)$ and $5\% (l = 2)$ [54], allowing for excitation of
predictions. Because the rotational constant of such another ground state Λ-doublet (NO) molecule. Long-tion and collision of a Rydberg molecule (here NO) with of ultralong-range molecules, comprised of the interac-
thermal molecular gas, rotational states are Boltzmann via Sisyphus cooling techniques [57], respectively. In a
within the realm of possibility with realistic microwave
state of NO
\[ f \]
Ref. [52], the NO(4
\[ f \]) state is an intermediary state to NO(n \text{g}) Rydberg levels.

In summary, we describe the formation of a new class of ultralong-range molecules, comprised of the interaction and collision of a Rydberg molecule (here NO) with another ground state A-doublet (NO) molecule. Long-lived Rydberg bi-molecular states with enormous PEDM are predicted. Because the rotational constant of such Rydberg bi-molecules are in the 0.1 – 1 MHz range, microwave spectroscopy of rotational transitions are now within the realm of possibility with realistic microwave Rabi frequencies. Cold and ultracold NO samples could be made in a supersonic beam expansion [53, 56] and via Sisyphus cooling techniques [57], respectively. In a thermal molecular gas, rotational states are Boltzmann distributed, but yet it is possible for the main rotational transition to be observed. Because the Rydberg electron couples to the spin symmetries in the molecule, the interaction terms in Eq. (2) can be spin-mixed and BO potentials with mixed spin and orbital angular momenta emerge, which can be manipulated and controlled with small magnetic fields. With a proliferation of activities in trapping of doublet molecules, in particular in optical tweezers [58] and magnetic traps [59], Rydberg excitation in such molecules in cold and ultracold settings is within reach. The ultralong-range Rydberg bi-molecules predicted in this work hence open a new vista into cold bi-molecular collisions and control interactions.

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