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Ultracold dipolar molecules composed of strongly magnetic atoms

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In a combined experimental and theoretical effort, we demonstrate a novel type of dipolar system made of ultracold bosonic dipolar molecules with large magnetic dipole moments. Our dipolar molecules are formed in weakly bound Feshbach molecular states from a sample of strongly magnetic bosonic erbium atoms. We show that the ultracold magnetic molecules can carry very large dipole moments and we demonstrate how to create and characterize them, and how to change their orientation. Finally, we confirm that the relaxation rates of molecules in a quasi-two dimensional geometry can be reduced by using the anisotropy of the dipole-dipole interaction and that this reduction follows a universal dipolar behavior.

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Ultracold dipolar particles are at the heart of very intense research activities, which aim to study the effect of interactions that are anisotropic and long range [1, 2]. Dipolar quantum phenomena require ultracold gases and a strong dipole-dipole interaction (DDI). So far, strongly dipolar gases have been obtained using either atoms with a large magnetic dipole moment or ground-state polar molecules with an electric dipole moment [2]. With both systems, many fascinating many-body quantum effects have been observed and studied, such as the d -wave collapse of a dipolar Bose-Einstein condensate [3, 4], the deformation of the Fermi sphere [5], and the spin-exchange phenomena [6, 7].

Here, we introduce a novel kind of strongly dipolar particles. These are weakly bound dipolar molecules produced from a pair of atoms with large magnetic dipole moments, such as erbium (Er). The central idea is that these molecules can possess a very large magnetic moment μ up to twice that of atoms (e.g. 14 Bohr magneton, μ_B , for Er_2) and have twice the mass of the atoms. As a consequence, the degree of “dipolarity” of the magnetic molecules is much larger than the one of atoms. This can be quantified in terms of the dipolar length $a_d = m\mu_0\mu^2/(4\pi\hbar^2)$ [1], which solely depends on the molecular mass m and on μ ; \hbar is the Planck constant divided by 2π . To give an example, Er_2 with $\mu = 14\mu_B$ has a a_d of about $1600a_0$, which largely exceeds the typical values of the s -wave scattering length. Here, a_0 is the Bohr radius. Moreover, in contrast to ground-state heteronuclear molecules, the dipole moment of the magnetic molecules does not vanish at zero external (magnetic) field, opening the intriguing possibility of investigating the physics of unpolarized dipoles.

In a joined experimental and theoretical effort, we study the key aspects of ultracold dipolar Er_2 molecules,

including the association process, the molecular energy spectrum, the magnetic dipole moments, and the scattering properties in both three- (3D) and quasi two-dimensional (q2D) geometries.

Erbium belongs to the class of strongly magnetic lanthanides, which are currently attracting great attention in the field of ultracold quantum gases [4, 8–10]. Indeed, these species exhibit unique interactions. Beside the long-range magnetic DDI, these species have both an isotropic and an anisotropic contribution in the short range van der Waals (vdW) potential. The latter results from the large non-zero orbital momentum quantum number of the atoms [11, 12]. This manifold leads to an extraordinary rich molecular spectrum, reflecting itself in a likewise dense spectra of Feshbach resonances as demonstrated in recent scattering experiments [4, 13, 14]. Each resonance position marks an avoided crossing between the atomic scattering threshold and a molecular bound state, which can be used to associate molecules from atom pairs [15].

We create and probe Er_2 dipolar molecules by using standard magneto-association and imaging techniques [15]. Details of the production schemes are described in the Supplemental Material [16]. In brief, we begin with an ultracold sample of ^{168}Er atoms in an optical dipole trap (ODT) in a crossed-beam configuration. The atoms are spin-polarized into the lowest Zeeman sublevel ($j = 6$, $m_j = -6$). Here, j is the atomic electronic angular momentum quantum number and m_j is its projection on the quantization axis along the magnetic field. To associate Er_2 molecules, we ramp the magnetic field across one of the low-field Feshbach resonances observed in Er [4, 13]. We experimentally optimize the ramping parameters, such as the ramp speed and the magnetic-field sweep interval, by maximizing the conver-

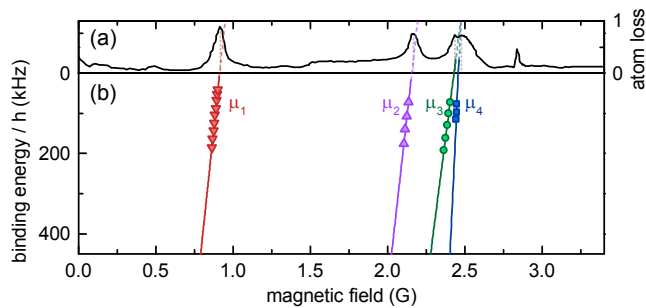


Figure 1. (color online). Er_2 weakly bound molecules. (a) Atom-loss spectrum [4] from 0 to 3 G and (b) near-threshold binding energy of the corresponding molecular states. The solid lines are fits to the experimental data and extrapolated to larger E_b up to $h \times 500$ kHz. The error bars are smaller than the symbols.

sion efficiency. In our experiment we typically achieve a conversion efficiency of 15%, which is a common value for boson-composed Feshbach molecules [15]. To obtain a pure molecular sample, we remove all the remaining atoms from the ODT by applying a resonant laser pulse. Our final molecular sample contains about 2×10^4 Er_2 Feshbach molecules at a temperature of 300 nK and at a density of about $8 \times 10^{11} \text{ cm}^{-3}$ [16].

A central question regards the magnitude of the dipole moment owned by the magnetic molecules. We experimentally determine μ by using magnetic-field modulation spectroscopy, a technique which was successfully applied to alkali atoms [17–19]. With this method, we measure the molecular binding energy E_b near the atomic threshold as a function of the magnetic field B . The binding energy is related to the differential magnetic moment of the molecules with respect to the atom-pair magnetic moment $2\mu_a$. Here, $\mu_a = -gm_j\mu_B = 6.98\mu_B$ in the case of Er, where $g = 1.16$ is the Er atomic Landé factor. We thus extract μ by using the relation $\mu = 2\mu_a - |dE_b(B)/dB|$. Our spectroscopic measurement begins with an ultracold atomic sample near a Feshbach resonance. We then add a small sinusoidal modulation to the bias magnetic field for 400 ms. The modulation frequency is varied at each experimental run. When it matches E_b/h , prominent atom losses appear because of molecule formation. We trace the near-threshold molecular spectrum by repeating the measurement for various magnetic-field values. Figure 1 shows the Er_2 molecular spectrum in a magnetic field range up to 3 G. In our range of investigation, we identify four molecular energy levels, which near threshold exhibit a linear dependence on B . For each state, we obtain a different μ value, ranging from 8 to $12\mu_B$ [20], as listed in Table I.

For alkali-metal atoms, which possess much simpler interaction properties than lanthanides, theoretical approaches based on coupled-channel calculations have been extremely successful in assigning the quantum num-

Table I. Experimental and theoretical magnetic moments of four molecular states near the atomic threshold, Feshbach-resonance positions B_{FR} , dipolar lengths, outer turning points R^* , and dominant quantum numbers ℓ , J , and M . For convenience, the molecular states are labeled as μ_i with $i = 1, \dots, 4$. The specified uncertainties correspond to the 1σ statistical errors.

	B_{FR} (G)	μ/μ_B		a_d (a_0)	R^* (a_0)	$ \ell, J, M\rangle$
		Exp.	Theo.			
μ_1	0.91	11.30(7)	11.20	1041(13)	72.0	$ 4, 12, -12/-10/-9\rangle$
μ_2	2.16	11.51(4)	11.46	1080(8)	71.0	$ 4, 10, -10\rangle$
μ_3	2.44	11.84(2)	11.75	1143(4)	86.0	$ 2, 12, -10\rangle$
μ_4	2.47	7.96(3)	7.92	517(4)	57.0	$ 6, 10, -7/-6\rangle$

bers of the molecular energy levels and reproducing molecular spectra [15]. However, a straightforward extension of these methods to the lanthanide case is out of reach because of their complex scattering physics involving highly anisotropic interactions and many partial waves [13]. Inspired by work on alkali-metal collisions [21–24], we develop a new theoretical approach to identify the molecular quantum numbers, based on approximate adiabatic potentials and on the experimentally measured μ as input parameters. Our scattering model is detailed in the Supplemental Material [16], whereas we here summarize the central ideas of our approach.

We first solve the eigenvalue problem of the full atom-atom interaction potential operator [16], whose eigenvalues are the adiabatic potentials $U_n(R; B)$. The corresponding eigenfunctions read as $|n; R\rangle = \sum_i c_{n,i}(R)|i\rangle$, where $n = 1, 2, \dots$ and $c_{n,i}(R)$ are R -dependent coefficients. The molecular state $|i\rangle$ is uniquely determined by the set of angular momentum quantum numbers (ℓ, J, M) , where ℓ is the molecular orbital quantum number, $\vec{J} = \vec{j}_1 + \vec{j}_2$ the total atomic angular momentum, and M its projection on the internuclear axis.

To derive the corresponding “adiabatic” molecular magnetic moments, we calculate $\mu^{\text{calc}} \approx -dU_n(R; B)/dB$ at the position of the outer classical turning point $R = R^*$. This choice is justified by the fact that most of the vibrational wavefunction is localized around R^* .

From the Hellmann-Feynman theorem it then follows that $\mu^{\text{calc}} = -g\mu_B \sum_i M_i |c_i(R^*)|^2$. Finally, we assume that for each Feshbach resonance a vibrational state is on resonance and we find the adiabatic potential that has a magnetic moment closest to the measured one within 1%. Once the best match is identified, the corresponding $|n; R\rangle$ sets the molecular state $|i\rangle$, characterized by ℓ , J , and M , with the largest, dominant contribution. In our range of investigation we observe d -, g -, and i -wave molecular states; see Table I. These states show several dominant M contributions. This fact is unusual and reflects the dominant role of the DDI, which couples several adiabatic potentials and M components. As shown in Fig. 2, this mixing effect is particularly dom-

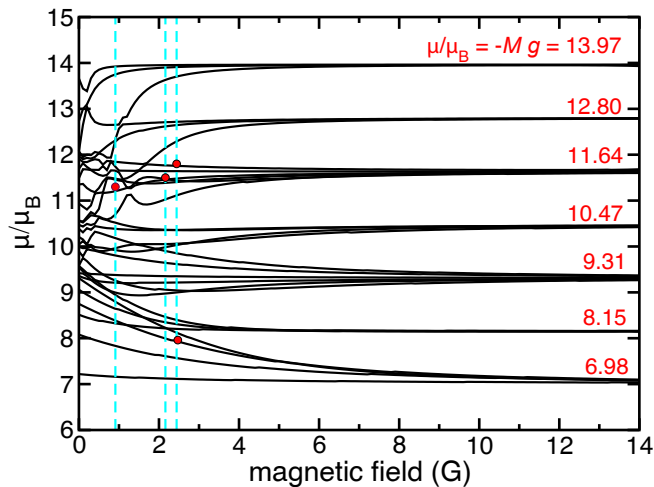


Figure 2. (color online). Adiabatic magnetic moment as a function of magnetic field strength evaluated at the entrance channel energy. Each curve corresponds to the adiabatic magnetic moment of one adiabatic potential $U_n(R; B)$. The magnetic moments in the asymptotic limit of large B are given. The dashed vertical lines correspond to the field strength where we have observed Feshbach resonances. The red-filled circles represent the experimentally measured magnetic moments at these resonance locations.

inant below 10 G, where the DDI at R^* is larger than the Zeeman interaction. Above 10 G, we predict μ to be equal to integer multiples of $g\mu_B$ [16].

As summarized in Table I, we find very good overall agreement between the measured and the calculated molecular magnetic moments. For the largest observed μ , we calculate a corresponding dipolar length $a_d \approx 1150 a_0$. This value exceeds the typical range of the vdW potentials, setting the DDI as the dominant interaction in the system. Remarkably, a_d for Er_2 is comparable to the one realized with ground-state KRb molecules [25], which are an extensively investigated case serving as a benchmark dipolar system.

Following the methods introduced for KRb [26–28], we test the dipolar character of Er_2 by performing scattering experiments in a three- (3D) and in a quasi two-dimensional (q2D) optical dipole trap. We control the DDI between molecules by tuning the dipole orientation, which is controlled by changing the direction of the magnetic field and is represented by the angle θ between the magnetic field axis and the gravity axis. Our experiment begins with the atomic sample trapped either in a 3D or in a q2D ODT. The q2D trap is created by superimposing a vertically oriented, one-dimensional optical lattice [16]. After the magneto-association and the removal of the remaining atoms, we probe the number of molecules as a function of the holding time in the ODT. We perform measurements for the molecular states μ_1 , μ_2 , and μ_4 [29]. For each of these states, we measure the collisional stability of the sample for both in-plane ($\theta = 90^\circ$) and

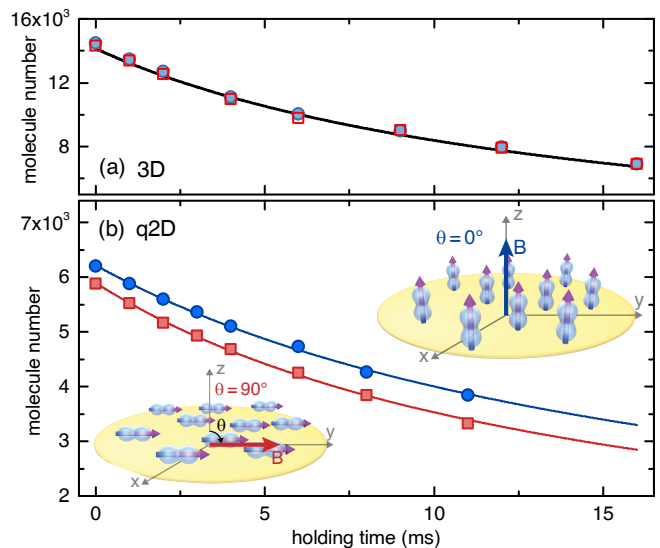


Figure 3. (color online). Typical time evolution of the number of molecules for $\theta = 90^\circ$ (squares) and $\theta = 0^\circ$ (circles) in a 3D (a) and in a q2D trap (b). The data refer to molecules in the state μ_1 for the 3D case (a) and molecules in the state μ_2 in q2D (b). The insets in (b) show an illustration of molecules in pancake-shaped traps with out-of-plane (right) and in-plane (left) orientations. The solid lines are two-body decay fits to the data. The error bars for (a) and (b) are smaller than the data points and are not shown. The data points in (a) are obtained by averaging 5 independent measurements and in (b) about 50 measurements have been averaged.

out-of-plane ($\theta = 0^\circ$) dipole orientation, and extract the corresponding relaxation rate coefficients, β_\perp and β_\parallel , using a standard two-body rate equation [30].

Figure 3 shows typical molecular decay curves in (a) 3D and in (b) q2D. In 3D, we confirm that the inelastic decay does not depend on θ . We obtain $\beta_{3D} = 1.3(2) \times 10^{-10} \text{ cm}^3/\text{s}$. This is a typical value for boson-composed Feshbach molecules, which undergo a rapid vibrational quenching into lower-lying molecular states, as demonstrated with alkali atoms [30]. Contrary, in q2D the decay rates clearly depend on the dipole orientation. For each investigated molecular state, β_\perp is larger than β_\parallel . We find a reduction of losses of up to 30% for out-of-plane orientation, for which the DDI is predominantly repulsive. The ratio $\frac{\beta_\perp(T)}{\beta_\parallel(T)}$ increases with increasing μ ; see Table II. We note that stronger suppression of losses can be obtained using a tighter two-dimensional confinement [26], which is presently not reachable with our experimental parameters.

The reduction of losses in q2D draws a natural analogy with the observations obtained with KRb molecules [28]. From a comparative analysis between Er_2 and KRb , one can unveil universal behaviour attributed to the DDI, for systems being different in nature, but sharing a similar degree of dipolarity. We thus theoretically study the scattering behavior of Er_2 using a theoretical approach

Table II. Experimental and theoretical loss rate coefficients β for $T = 400$ nK and for various μ and θ at $B = 200$ mG. Uncertainties of β are statistical from fitting and systematic due to number density uncertainty. For the slightly different values of μ compared to Table I and the error discussion see Supplemental Material [16].

	μ/μ_B	β_{\perp} (10^{-6} cm ² /s)		β_{\parallel} (10^{-6} cm ² /s)	
		Exp.	Theo.	Exp.	Theo.
μ_4	8.7(6)	$12.5 \pm 0.3 \pm 3.3$	6.00	$10.6 \pm 0.3 \pm 2.8$	4.79
μ_1	10.9(5)	$9.5 \pm 0.2 \pm 2.5$	6.81	$7.3 \pm 0.1 \pm 2.1$	5.07
μ_2	11.7(3)	$11.3 \pm 0.2 \pm 2.9$	7.12	$8.6 \pm 0.2 \pm 2.3$	5.13

similar to the one successfully applied to KRB. Our formalism, which accounts for the DDI and the isotropic vdW interaction, is described in Refs. [16, 31].

We compute the $\text{Er}_2 + \text{Er}_2$ loss rate coefficients $\beta(T)$ in 3D and in q2D for given values of μ , θ , and T . By averaging over a 3D and a 2D Maxwell-Boltzmann distribution, we obtain the thermalized loss rate coefficients $\beta(T)$ in 3D and in q2D, respectively. In 3D, we find a rate coefficient of 1.0×10^{-10} cm³/s at $T = 300$ nK, which is close to the experimental value [32]. In q2D, our calculations show that the collision dynamics at long range, and thus the value of β , depends on the dipole orientation and monotonically increases with μ . As in the experiments, our calculations show that collisions for in-plane orientation (β_{\perp}) lead to larger molecular losses than for out-of-plane orientation (β_{\parallel}). In Table II, we compare theory and experiment. The absolute values of β agree within a factor of two. This difference is well explained by the fact that our model does not include details of the short-range physics, being the Er_4 potential energy surfaces currently unknown [16].

Remarkably, the experimental and calculated ratios $\frac{\beta_{\perp}(T)}{\beta_{\parallel}(T)}$ agree very well with each other; see Fig. 4. This suggests that $\frac{\beta_{\perp}(T)}{\beta_{\parallel}(T)}$ for Er_2 Feshbach molecules is determined by the DDI and not by the short-range physics, and that it can be correctly described using a point-like-dipole formalism [16]. Figure 4 shows the comparative analysis between bosonic $^{41}\text{K}^{87}\text{Rb}$ and $^{168}\text{Er}_2$, and fermionic $^{40}\text{K}^{87}\text{Rb}$ and $^{167}\text{Er}^{168}\text{Er}$ based on our numerical calculations. Independent of the nature of the magnetic or electric dipolar system, we find universal curves as a function of a_d/\tilde{a} : one for bosons with $\tilde{a} = a_{\text{ho}}$ and one for fermions when $\tilde{a} = a_{\text{vdW}}$. Here, a_{ho} is the harmonic oscillator length and $a_{\text{vdW}} = (2mC_6/\hbar^2)^{1/4}$ is the vdW length with C_6 the vdW coefficient. The faster increase of $\frac{\beta_{\perp}}{\beta_{\parallel}}$ for fermions with respect to bosons is due to the statistical fermionic suppression of β_{\parallel} in q2D that does not occur for bosons as explained in Ref. [33].

The universal behavior of ultracold dipolar scattering has been previously pointed out in Ref. [34]. In the Wigner regime, we derive simple universal scaling laws for dipolar bosonic and fermionic molecules [16, 34]. For

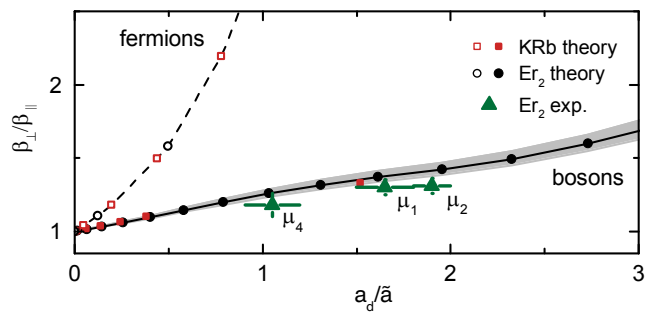


Figure 4. (color online). Universal loss rate ratio $\beta_{\perp}/\beta_{\parallel}$ as a function of a_d/\tilde{a} for Er_2 (circles) and KRB (squares) for a fixed value $a_{\text{dB}}/a_{\text{ho}} = 4.85$ corresponding to $T = 400(40)$ nK and $\nu_z = 31.2(1)$ kHz [16]. The gray shaded area is due to the uncertainty of T . Here, $\tilde{a} = a_{\text{ho}}$ for bosonic molecules (filled symbols) and $\tilde{a} = a_{\text{vdW}}$ for fermionic molecules (open symbols). The calculated loss rate ratios of Er_2 are compared with the experimental data for states μ_1 , μ_2 , and μ_4 (triangles).

bosons with $a_d, a_{\text{ho}} > a_{\text{vdW}}$, which is the case of our Er_2 molecules, we find $\frac{\beta_{\perp}(T)}{\beta_{\parallel}(T)} \sim \left(\frac{a_{\text{dB}}}{a_{\text{ho}}}\right)^4 \frac{a_d}{a_{\text{ho}}} \exp\left[2\left(\frac{a_d}{a_{\text{ho}}}\right)^{2/5}\right]$. For fermions with $a_d, a_{\text{vdW}} < a_{\text{ho}}$, $\frac{\beta_{\perp}}{\beta_{\parallel}} \sim \left(\frac{a_d}{a_{\text{vdW}}}\right)^3$. Here, $a_{\text{dB}} = h/\sqrt{2\pi mk_B T}$ is the thermal de Broglie wavelength.

To conclude, our work reports on the study of strongly dipolar molecules created by pairing ultracold atoms with large magnetic dipole moments. We anticipate that our scheme can be generalized to other magnetic lanthanide species and has the potential to open regimes of investigations, which have been inaccessible so far. First, the extraordinarily dense and rich molecular energy spectrum of Er opens the exciting prospect of cruising through molecular states of different magnetic moments or even creating molecular-state mixtures with dipole imbalance [19, 35, 36]. Second, in contrast to electric polar molecules where the electric dipole moment is zero in the absence of a polarizing electric field, magnetic dipolar molecules have a permanent dipole moment allowing to study the physics of unpolarized dipoles. In addition, strongly magnetic Feshbach molecules offer a novel case of study for scattering physics. These molecules are in fact diffuse in space with a typical size on the order of the vdW length. This novel situation can also have interesting consequences and trigger the development of extended scattering models, which account for multipolar effects and truly four-body contributions when the molecule size becomes comparable to a_d [37]. Finally, a very promising development will be to create fermionic Er_2 dipolar molecules where vibrational quenching processes are intrinsically suppressed because of the Pauli exclusion principle [38, 39].

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