Metastability is at the heart of many phenomena in physics. Energy barriers or conservation laws can efficiently prevent a system from decaying into lower-lying states. In the field of ultracold quantum gases, metastability is ubiquitous. Bose-Einstein condensates in dilute alkali vapors are intrinsically metastable, as the absolute stability is ubiquitous. Bose-Einstein condensates in dilute states. In the field of ultracold quantum gases, metastability is ubiquitous. Bose-Einstein condensates in dilute alkali vapors are intrinsically metastable, as the absolute ground state of the system is just a tiny piece of metal. Further prominent examples for the important role of metastability in ultracold gases can be found in Bose-Einstein condensates with attractive interactions \cite{1, 2}, vortices in rotating superfluids \cite{3, 4}, dark solitons \cite{5, 6}, and repulsively bound atom pairs in optical lattices \cite{7, 8}.

The association of Feshbach molecules in ultracold quantum gases \cite{8} has opened up many new opportunities in the field, e.g. for experiments on quantum states with strong pair correlations. It has become experimental routine to produce dimers in a binding energy range of typically 100 kHz up to a few ten MHz below the atomic threshold. Considerable efforts are in progress to extend the energy range of ultracold molecules initially prepared by Feshbach association to larger binding energies \cite{9, 10}, ultimately with the goal to produce quantum-degenerate molecular samples in the rovibrational ground state.

In this Letter, we explore ultracold molecules in a new energy regime. We create metastable dimers above the threshold for spontaneous dissociation into free atoms, i.e. dimers with negative binding energies. In many experiments, the fast dissociation of Feshbach molecules above threshold on the time scale of microseconds \cite{11, 12} is used for detection purposes by imaging the resulting atoms. Feshbach molecules with energetically open channels for spontaneous dissociation were investigated in two previous experiments. Lifetimes of up to a few tens of milliseconds were demonstrated in an \( s \)-wave halo state of \(^{85}\text{Rb}_2 \) \cite{13} and, more recently, \(^{40}\text{K}_2 \) \( p \)-wave molecules above threshold behind a centrifugal barrier were observed with lifetimes of a few hundreds of microseconds \cite{14}. In contrast to these rather limited lifetimes, we demonstrate above-threshold molecules that are stable against dissociative decay on the time scale of a second; this can be considered infinitely long for experimental purposes. The basic idea to achieve such metastability is the transfer of the dimers to a high rotational state where a large centrifugal barrier in combination with a weak coupling to low partial-wave scattering states strongly suppresses spontaneous decay; see illustration in Fig. 1. Our experiments are performed on ultracold, trapped \(^{133}\text{Cs}_2 \) dimers transferred into a state with rotational quantum number \( \ell = 8 \), i.e. into an \( l \)-wave state \cite{15}; here the centrifugal energy barrier is as high as \( h \times 150 \text{MHz} \).

The two-body scattering properties of Cs and the underlying molecular structure have been thoroughly investigated in previous work \cite{16, 17}. Feshbach resonances up to \( g \)-wave character have been found. The underlying coupling of the \( s \)-wave scattering continuum to \( g \)-wave molecular states is special to cesium because of relatively strong indirect spin-spin coupling \cite{18}. This second-order
to the scattering continuum for atoms, which is a consequence of the negligible coupling. Remarkably, these states do not manifest themselves above the dissociation threshold \((E = 0)\) and the height of the \(d\)-wave centrifugal barrier \((E = h \times 3.7 \text{ MHz})\) are indicated. The quantum state relevant for this work is labeled with \(6l(4)\). The open squares represent spectroscopic data on this state obtained from measuring its magnetic moment, and the arrows indicate the path for preparation of the dimers (for details see text).

interaction in general allows coupling between different partial-wave states with a difference \(\Delta\ell\) in rotational quantum numbers up to \(|\Delta\ell| = 4\). The Cs dimer energy structure in the relevant range is shown in Fig. 2. The \(s\)-wave threshold corresponds to two free atoms in the lowest hyperfine ground state sublevel \(|F, m_F| = |3, 3\rangle\) with zero kinetic energy. The notation \(f\ell(m_F)\) for molecular states was introduced in Ref. \([17]\); the symbols \(f\) and \(m_F\) represent the quantum numbers for the total internal angular momentum and its projection.

The CsCs spectrum provides several accessible \(l\)-wave states, which have been identified in Refs. \([17, 19, 20]\). Remarkably, these states do not manifest themselves in observable Feshbach resonances in the scattering of atoms, which is a consequence of the negligible coupling to the scattering continuum for \(|\Delta\ell| > 4\). The \(l\)-wave states can nevertheless be populated efficiently by using coherent state-transfer at avoided level crossings \([17]\), where each step obeys the selection rule \(\Delta\ell \leq 4\) for the rotational angular momentum.

In this work, the state \(6l(4)\) serves as a model system for metastable dimers in high rotational states above the dissociation threshold. The starting point for our experiments is a sample of typically \(1 \times 10^4\) \(l\)-wave dimers prepared in a crossed-beam CO\(_2\) laser trap at a temperature of \(T = 250(50)\) nK; the mean trap frequency is \(\bar{\omega} = 2\pi \times 41(1)\) Hz. The experimental procedures have been developed earlier \([17]\). In brief, molecules are first created by Feshbach association in the state \(4g(4)\) using the 19.84-G resonance, followed by transfer into \(6l(4)\) via an intermediate state \(6g(6)\); the latter state has a binding energy of about \(h \times 5\text{ MHz}\) and is out of the energy range displayed in Fig. 2. The state \(6l(4)\) has a magnetic moment of \(0.96(1)\mu_B\) and crosses the dissociation threshold at a magnetic field of 22.0(2) G. We have spectroscopically investigated this state by extending magnetic-moment measurements \([17]\) into energy regions above threshold; see open squares in Fig. 2.

To measure the lifetime of the \(l\)-wave state, we hold the sample at magnetic fields corresponding to energies above and below the dissociation threshold. After variable storage times of up to 0.4 s, the remaining molecules are quickly (within typically 10 ms) subjected to the reverse transfer route and dissociated at the \(g\)-wave Feshbach resonance at 19.84 G. Standard absorption imaging is finally applied to the resulting atom sample \([21]\).

In Fig. 3 we show typical decay measurements. Just below threshold \((B = 21.6\text{ G},\) filled squares), we observe a non-exponential decay of the molecular sample on a timescale of 100 ms, which is a result of inelastic two-body collisions. For an energy of \(h \times 2\text{ MHz}\) above threshold \((24.7\text{ G},\) open triangles), the behavior is very similar with a slightly faster decay. Only at magnetic fields around 27 G we observe a much faster loss of molecules. We analyze the decay measurements using the rate equation \(\dot{N}/N = -\alpha - L_2\bar{n}\), where \(L_2\) is the loss coefficient that describes two-body decay resulting from inelastic dimer-dimer collisions. The parameter \(\alpha\) represents the rate of spontaneous dissociation as the dominant one-body decay process of the dimers. The mean molecu-

FIG. 2: (Color online) Energy curves of the relevant Cs\(_2\) dimer states as function of the magnetic field \([16, 17]\). The \(s\)-wave threshold \((E = 0)\) and the height of the \(d\)-wave centrifugal barrier \((E = h \times 3.7\text{ MHz})\) are indicated. The quantum state relevant for this work is labeled with \(6l(4)\). The open squares represent spectroscopic data on this state obtained from measuring its magnetic moment, and the arrows indicate the path for preparation of the dimers (for details see text).

FIG. 3: (Color online) Loss measurements of the \(6l(4)\) molecular sample for different magnetic fields. One set of data is taken below the dissociation threshold \((B = 21.6\text{ G})\); three further sets \((24.7\text{ G}, 27.0\text{ G}, 27.1\text{ G})\) refer to situations above threshold. The lines are fits to the measurements including two-body and one-body decay. The inset shows the resulting two-body loss coefficient \(L_2\), where the shaded region indicates the magnetic field region above threshold. The horizontal line indicates the average \(L_2\) below threshold.
lar density \( \bar{n} \) is related to the molecule number \( N \) by
\[
\bar{n} = \frac{m \omega^2}{(2 \pi k_B T)^{3/2}} N,
\]
where \( m \) is the mass of a Cs atom. Below threshold the loss of molecules is solely determined by inelastic two-body decay and the one-body decay term can be omitted. Other loss sources, such as light-induced dissociation and background collisions, can be neglected under our experimental conditions.

The fit results for \( L_2 \) as a function of the magnetic field are shown in the inset of Fig. 4. Below threshold no magnetic field dependence is observed. Here \( L_2 \) has a value of 4.2 \((0.2)_{\text{stat}}(1.4)_{\text{syst}} \times 10^{-11} \text{ cm}^3 \text{s}^{-1} \), similar to the values for \( \text{Cs}_2 \) found in other quantum states [19, 22]. The systematic error is based on the uncertainty in the temperature and the trap frequency measurements. Above threshold \( \alpha \) is a free parameter in the fit procedure and the competition between one-body and two-body decay introduces a large error. The values obtained for \( L_2 \) above threshold scatter around the value obtained below threshold with a trend to somewhat higher values.

The results for the dissociation rate \( \alpha \) are shown in Fig. 4 (filled symbols). We also apply a constrained fit, in which \( L_2 \) is fixed to its constant value below threshold. The results of this constrained fit (open symbols) give an upper limit for the dissociation rates under the plausible assumption that collisional decay does not decrease above threshold. Below 26 G, i.e. up to 4 G above threshold, the dissociation rates remain close to zero. Around 27 G, however, an increase is observed, which is most clearly seen from the constrained fit. One-body lifetimes for spontaneous dissociation in the regime of negative binding energies above threshold are obtained by inverting the dissociation rates and are shown in the inset of Fig. 4 as function of the energy above threshold. The one-body lifetimes up to 26 G \(( h \times 3 \text{ MHz} \) are at least 1 s, which highlights the long-lived metastable character of the \( l \)-wave Feshbach molecules. Beyond this time scale, we cannot rule out a very slow dissociative decay, e.g. by a high-order coupling to the \( s \)-wave scattering continuum.

The increase of the dissociation rate around 27 G shows that a new dissociation channel opens up. This behavior cannot be explained by tunneling through the \( l \)-wave centrifugal barrier, as the estimated rate for this process is four orders of magnitude smaller than observed here experimentally. We explain the onset of dissociation by coupling to a \( g \)-wave state, which mediates the dissociation [23]. The \( 6l(4) \) state crosses a state \( 6g(5) \) at about 28 G (see Fig. 2). When the crossing is approached, a \( g \)-wave component starts to mix in. This increasing admixture then allows decay into \( s \) - and \( d \)-waves without the requirement of any higher-order coupling beyond the indirect spin-spin interaction.

To investigate the dissociation process around 27 G in more detail, we directly image the resulting atoms [14, 24], without the reverse transfer route and dissociation at 19.84 G. In Fig. 5 we show the number of dissociated molecules as function of the magnetic field. Here the images are taken 5 ms after reaching the final magnetic field value in a fast ramp. Below 26 G no dissociated molecules are observed, while in the region of 27-28 G a rapid appearance is seen. The inset of Fig. 5 shows an absorption image at 27.8 G. The dissociation pattern is consistent with an interference between \( s \) - and \( d \)-waves. The presence of \( d \)-waves is expected as the dissociation occurs above the \( d \)-wave centrifugal barrier.

We use a simple two-channel avoided crossing approach to model the magnetic field dependence of the dissociation rate. Near the crossing, we describe the molecular...
state as a superposition of the $l$- and $g$-wave states. The $g$-wave amplitude determines the dissociation rate and increases smoothly from zero to one when the avoided crossing of quasi-bound states is followed from below. A fit based on this model is shown by the solid line in Fig. 2. The model allows us to determine the position of the crossing within an uncertainty of 0.1 G. We obtain a value of 27.7 G, which is consistent with our previous knowledge of the molecular spectrum [17].

The dissociation mechanism discussed for the 6$l$(4) state also agrees with observations in further experiments on the two other neighboring $l$-wave states 6$l$(3) and 6$l$(5). The latter state crosses the atomic threshold at 35.0(2) G and has a first avoided crossing with a $g$-wave state only around $h \times 12$ MHz (not shown in Fig. 2). Here we explored magnetic fields up to 48 G and corresponding energies up to $h \times 8$ MHz without finding any significant decay, even far above the $d$-wave centrifugal barrier [23]. In contrast, the 6$l$(3) state shows significant dissociation already at a relatively small energy of $h \times 0.7$ MHz above threshold [20]. This is explained by the presence of a crossing with the 6$g$(5) state, which is found at a magnetic field only 0.5 G higher than the intersection of the 6$l$(3) state with the $s$-wave threshold (see Fig. 2).

To conclude, we have demonstrated the metastable character of ultracold dimers in a high rotational state above the dissociation threshold, where the binding energy is negative. In our experiments, we observed Cs$_2$ Feshbach molecules in an $l$-wave state to be stable against dissociative decay on a time scale of at least one second. The large centrifugal barrier suppresses tunneling to the $l$-wave scattering continuum while the coupling to lower partial waves is extremely small. The metastable region and onset of dissociation for the $l$-wave states depends on the location of the first crossing with a $g$-wave state, which mediates dissociative decay.

In future work, shielding the dimers against collisional decay can be achieved by trapping in an optical lattice [26]. In such a periodic environment one may also create a Mott-like state with exactly one molecule per lattice site [27]. This opens up the possibility to create novel metastable quantum states with strong pair correlations. The possibility to achieve dissociative decay on demand with a well-defined energy by switching the magnetic field to particular values opens up further possibilities, e.g. for a controlled collective decay of the inverted medium in analogy to superradiance.

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