Many-body chemical reactions in a quantum degenerate gas

Chemical reactions in the quantum degenerate regime are described by the mixing of matter-wave fields. In many-body reactions involving bosonic reactants and products, such as coupled atomic and molecular Bose–Einstein condensates, quantum coherence and bosonic enhancement are key features of the reaction dynamics. However, the observation of these many-body phenomena, also known as ‘superchemistry’, has been elusive so far. Here we report the observation of coherent and collective reactive coupling between Bose-condensed atoms and molecules near a Feshbach resonance. Starting from an atomic condensate, the reaction begins with the rapid formation of molecules, followed by oscillations of their populations during the equilibration process. We observe faster oscillations in samples with higher densities, indicating bosonic enhancement. We present a quantum field model that captures the dynamics well and allows us to identify three-body recombination as the dominant reaction process. Our findings deepen our understanding of quantum many-body chemistry and offer insights into the control of chemical reactions at quantum degeneracy.

Ultracold atoms and molecules form an ideal platform towards controlling chemical reactions at the level of single internal and external quantum states. Ultracold molecules can be prepared in an individual internal state by, for example, the magnetoassociation and photoassociation of ultracold atoms and direct laser cooling. The external motion of molecules can be constrained by loading them into optical lattices or tweezers. These experiments lead to the realization of state-to-state ultracold chemistry. A number of experiments on cold molecules have reached the regime of quantum degeneracy, which promise new forms of molecular quantum matter and reaction dynamics. For instance, molecular Bose–Einstein condensates (BECs) formed in atomic Fermi gases have stimulated tremendous interest in the BEC-to-Bardeen–Cooper–Schrieffer crossover. Degenerate fermionic molecules are created by the magnetoassociation of bosonic and fermionic atoms and optical transitions to the lowest rovibrational state. Here quantum degeneracy suppresses chemical reactions due to the fermion antibunching effect.

Recently, molecular BECs have been realized based on atomic BECs near a Feshbach resonance. The reactive coupling between condensed atoms and molecules promises a new regime of quantum chemistry, dubbed ‘quantum superchemistry’, which highlights the coherent coupling of macroscopic matter waves and Bose stimulation of the reaction process. A key feature of this coherence is the collective oscillations between the reactant and product populations. Because of Bose statistics, the collective enhancement of reaction dynamics is anticipated near a Feshbach resonance.

At quantum degeneracy, the reaction dynamics fundamentally arise from the mixing of the matter-wave fields of the reactants and products. For instance, consider Feshbach coupling that converts two atoms into one molecule and vice versa, described by the chemical equation: \( \text{A + A} \leftrightarrow \text{A}_2 \). In a quantum gas, the reaction is described by the many-body Hamiltonian with the reaction order \( \alpha = 3 \):

\[
\hat{H} = \varepsilon_m \hat{\psi}_m^\dagger \hat{\psi}_m + g_2 \hat{\psi}_m^\dagger \hat{\psi}_m \hat{\psi}_2^\dagger \hat{\psi}_2,
\]

where \( \hat{\psi}_m (\hat{\psi}_n) \) is the atomic (molecular) field operator, \( g_2 \) is the Feshbach coupling strength and \( \varepsilon_m \) is the energy of one bare molecule relative to two bare atoms. Here we define the reaction order \( \alpha \) as the maximum number of field operators in reaction terms.

Another prominent example that couples ultracold atoms and molecules is three-body recombination, where three colliding atoms are converted into a diatomic molecule and another atom,

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and vice versa. This process is described by the chemical equation 
$A + A + A \rightarrow A_2 + A$. At quantum degeneracy, the recombination process can resonantly couple atomic and molecular fields as

$$\hat{H}_3 = g_3 \hat{\psi}_a^\dagger \hat{\psi}_a^\dagger \hat{\psi}_m \hat{\psi}_m + g_3 \hat{\psi}_a^\dagger \hat{\psi}_m \hat{\psi}_a \hat{\psi}_m,$$  

(2)

where $g_3$ is the recombination coupling strength. Here the reaction order is $\alpha = 5$.

To understand the dynamics of the coupled quantum fields, we present the following picture. We show that the molecular population $N_m = \hat{\psi}_m^\dagger \hat{\psi}_m$ follows the form of an ‘energy conservation’ law as

$$\frac{\hbar^2}{2} \frac{N_m^2}{N_m} + V(N_m) = \text{const.},$$  

(3)

where $\hbar^2 N^2_m/2$ resembles the kinetic energy and we introduce the many-body reaction potential $V = [N_m, \hat{H}]^2/2 + \text{const.}$ (Supplementary Information), which connects the reactants and products. In this picture, the system tends towards a lower potential. Quantum fluctuations of the nonlinear field coupling, however, can effectively damp the dynamics of the populations. In experiments, damping can also come from inelastic field coupling and coupling to a thermal field. Thus, one expects that the system first relaxes towards the potential minimum, and then equilibrates near the minimum with small-amplitude quantum fluctuations (Fig. 1). In the thermodynamic limit with total particle number $N \gg 1$, the reaction potential and oscillation frequency near the minimum scale with the particle number as $V \propto N^\alpha$ and $\omega_0 \propto N^{\alpha-1}$. The dependence on the particle number signals the bosonic enhancement of the reaction dynamics.

In this paper, we report the observation of coherent and Bose-stimulated reactions between Bose-condensed Cs atoms and Cs$_2$ molecules near an atomic Feshbach resonance. The reaction is initiated by tuning the magnetic field near a narrow $g$-wave Feshbach resonance, which couples scattering atoms and diatomic Feshbach molecules in a single high-lying rovibrational state bounded by van der Waals potential (Methods). Near the resonance, atomic and molecular populations quickly relax towards a dynamical equilibrium, followed by coherent oscillations between atoms and molecules in the equilibration process. We show that the oscillation frequency strongly depends on the particle number. From the dependence, we conclude that three-body recombination is the dominant reaction process that couples the atomic and molecular fields near the Feshbach resonance.

Our experiment starts with an ultracold Bose gas of $6 \times 10^3$ to $5 \times 10^5$ caesium atoms in an optical trap. The atoms can form a pure BEC either in a three-dimensional harmonic potential or a two-dimensional (2D) square well potential. We induce the reaction by switching the magnetic field near the $g$-wave Feshbach resonance, which can convert an atomic BEC into a molecular BEC. We determine the resonance position $B_0 = 19.849(2)$ G, resonance width $\Delta B = 8.3(5)$ mG and relative magnetic moment $\delta \mu = h \times 0.76(3) \text{ MHz} G$, where $h$ is the Planck constant, from the measurements of molecular binding energy $\varepsilon_m = \delta \mu (B - B_0)$ and the scattering length (Methods). After the reaction, we decouple the reaction potential $V$ above the Feshbach resonance at $B_0 = 19.849(2)$ G. The solid lines are fits to the data in the initial growth stage for the extraction of molecule formation rate $N_m$. Extracted molecule formation rate coefficient $\beta$ above and below the critical temperature $T_c$. The red line is a fit to the data based on the classical kinetic theory prediction $\beta = \beta_c \frac{\Gamma}{\Gamma_b}$, from which we obtain the classical branching ratio $\beta_c = 7(1)$% (see the main text). The blue line fits the data in the quantum regime with $\beta = \beta_q \frac{\Gamma}{\Gamma_b}$, which gives the quantum branching ratio $\beta_q = 3.9(3)$% (see the main text). The inset shows the rate coefficient normalized to the classical gas expectation $\epsilon_c$. In a, the error bars represent one standard deviation of the mean, estimated from 4–8 measurements. In b, the error bars represent 95% confidence intervals of the mean.
the atoms and molecules by quickly tuning the magnetic field far off the resonance and image each independently\(^1\). To show that chemical reactions follow different rules in a degenerate quantum gas versus a normal gas, we compare the molecule production and heating of both atoms and molecules. We identify this fast process as the relaxation dynamics (Fig. 1b). To characterize the enhanced reaction rate, we fit the loss rate near the resonance as

\[
\gamma_a = N_0 \langle \sigma v \rangle / N_0 T_a
\]

where \(N_0\) is the initial atomic density. We observe a steep drop in the rate coefficient \(\gamma_a\) at low temperatures, where we model the rate coefficient as

\[
\gamma_a \propto T_a^{-1/2}
\]

to fit the loss rate near the resonance as

\[
\gamma_a \propto T_a^{-\alpha}
\]

where \(\alpha = 0.5\) (Fig. 2b). At low temperatures, we model the rate coefficient as

\[
\gamma_a = b_0 T_a^\alpha
\]

where \(b_0\) and \(\Gamma_0 = 4\sqrt{3h/\hbar} m/(6n^2 \pi)\) are the branching ratio and rate coefficient predicted by the universal theory in the quantum regime, respectively\(^{20,21}\). The model fits the loss rate well, and we extract the branching ratio to be \(b_0 = 3.9(3)\). The sharp transition of the molecule formation rate around critical temperature \(T_c\) indicates different laws in the classical and quantum degenerate regimes. The suppression of the reaction rate in atomic BECs can be attributed to the suppression of correlations in the condensed phase\(^2\), as well as strong interactions between atoms and molecules across the Feshbach resonance.

A close examination of the molecule formation dynamics in atomic BECs reveals additional interesting features of quantum many-body reactions. To understand the underlying reaction processes, we study the atom loss rate \(\gamma_a = N_0 / N_0 T_a\) right after switching the magnetic field, where \(N_0\) is the atom number (Fig. 3c). Far from the resonance \(|B - B_0| \gg \Delta B\), atoms decay slowly and the loss rate follows a symmetric Lorentzian profile centred at the resonance \(\gamma_a = (B - B_0)^2\). We attribute the symmetric Lorentzian lineshape to the Feshbach coupling that off-resonantly couples two atoms to a molecule, and the molecule is lost when it collides with a third atom inelastically\(^2\).

Near the resonance, the loss rate greatly exceeds the expectation from the Lorentzian profile. This rapid atom loss only lasts for a few hundreds of microseconds and is accompanied with fast molecule production and heating of both atoms and molecules. We identify this fast process as the relaxation dynamics (Fig. 1b). To characterize the enhanced reaction rate, we fit the loss rate near the resonance as

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Fig. 4 | Bose-enhanced atom–molecule reaction dynamics on Feshbach resonance. a, Molecules formed in atomic BECs with different initial atom numbers \( N_0 \) following the magnetic field quench to 1(1) mG below the Feshbach resonance. The solid lines are fits to the data. b, Extracted oscillation frequencies for different initial atom numbers \( N_0 \). The red (magenta) solid line is a power-law fit with exponent given by the two-body (three-body) model. The blue dashed line is a power-law fit with a free exponent, which yields the scaling \( \omega_0 \propto N_0^{6/5} \). In the inset, we show the frequency dependence on the mean atomic density \( n_0 \) and the associated fits that yield the scaling \( \omega_0 \propto N_0^{6/5} \). c, Reaction potential \( V \equiv V_1 = -4g_1^2N_0^3f_m(1 - f_m)^3 \) of the three-body process described in equation (2) for different total particle numbers \( N \) (green solid lines). In the thermodynamic limit \( N \to 1 \), the minimum occurs at molecule fraction \( f_m = 2N_0/N = 1/5 \). d, Evolution of molecule fraction \( f_m \) for different initial atom numbers \( N_0 \). The mean molecule fractions in the equilibration phase \( f_m = 19(2)\% \) and \( f_m = 21(1)\% \) are consistent with the predicted minimum position of \( V_1 \) at \( f_m = 1/5 \). Here the uncertainties represent 95% confidence intervals. Data in a and d are averages of 3–5 measurements and the error bars represent one standard deviation of the mean.

\[
y_0 \propto [1 + (B - B_0)/(i\hbar)]^{-1}
\]
from which we extract the exponents \( c = 2.9(4) \) above the resonance and \( c = 6(2) \) below the resonance. The exponents \( c \), larger than 2 are consistent with the enhanced atom loss near the resonance beyond the Lorentzian profile.

The relaxation dynamics stem from three-body recombination, evidenced by the fast heating of both species in the relaxation phase (Fig. 3b). In addition, the measured exponent \( c = 2.9(4) \) from the enhanced atom loss is consistent with the predicted value of 3 for three-body recombination near a narrow Feshbach resonance.43,45 We attribute the even larger exponent \( c = 6(2) \) below the resonance to the bosonic enhancement of the three-body process.

Following the relaxation, both atomic and molecular populations oscillate for several milliseconds before they slowly decay over a much longer timescale (Fig. 3a, Methods). The oscillation indicates a coherent coupling between the atomic and molecular fields, consistent with the equilibration dynamics near the reaction potential minimum (Fig. 1b). The frequency \( \omega \) of oscillation depends on the magnetic field and is well fit to \( \omega = \sqrt{\Omega_0^2 + \omega_0^2} \) (Fig. 3d). Far from the resonance, the frequency approaches the molecular binding energy \( \epsilon_0 \). On resonance with \( \epsilon_0 = 0 \), the frequency \( \omega = \omega_0 \) is given by the collective reactive coupling between the atomic and molecular fields.

To demonstrate the many-body nature of the reactive coupling, we probe the atom–molecule oscillations right on the Feshbach resonance with different initial atom numbers \( N_0 \) and mean densities \( n_0 \). After quenching the magnetic field, we observe that samples with higher populations and densities display faster oscillations (Fig. 4a). Fitting the data, we obtain the scaling \( \omega_0 \propto N_0^{6/5} \) and \( \omega_0 \propto N_0^{1.7(4)} \) (Fig. 4b). Note that the two scalings are linked by \( n_0 \propto N_0^{1.5} \) for BECs in a harmonic trap.46,47 The particle number dependence of the reactive coupling supports the bosonic enhancement of the reaction process.

The scaling with respect to the particle number also reveals the underlying reaction mechanism. For the three-body recombination process described in equation (2), we derive the effective potential \( V_3 = -g_3^2N_0^3f_m(1 - f_m)^3 + O(N^4) \), where \( f_m = 2N_0/N \) is the molecule fraction, from which the resonant oscillation frequency is calculated to be \( \omega_0 \propto N^{6/5} \) in a harmonic trap (Supplementary Information). For the two-body recombination process described in equation (1), the effective potential is \( V_2 = -g_2^2N_0^2f_m(1 - f_m)^2 + O(N^3) \) which yields the scaling \( \omega_0 \propto N^{13/7} \). Our measurement agrees well with the three-body model (Fig. 4b).

Moreover, we find that the molecule fraction oscillates at around 20(1)% in the equilibration phase, which is consistent with the minimum position of the reaction potential \( V_1 \) at \( f_m = 1/5 \) (Fig. 4c, d). The two-body Feshbach process, on the other hand, predicts a different minimum of \( V_2 \) at \( f_m = 1/3 \). We note that the total population decays over a timescale longer than 5 ms, and thus, our theory model can describe the coherent atom–molecule coupling that occurs at a few kilohertz. The overall population loss due to inelastic collisions can contribute to damping of the coherent oscillations.

To conclude, we observe collective many-body chemical reactions in an atomic BEC near a Feshbach resonance. The dynamics are well described by a quantum field model derived from three-body recombinations. In particular, the coherent oscillations of atomic and molecular fields in the equilibration phase support quantum coherence and Bose enhancement of the reaction process. The observation of coherent and collective chemical reactions in the quantum degenerate regime paves the way to explore the interplay between many-body physics and ultracold chemistry.
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Methods
Experimental procedure
Our experiment starts with an ultracold Bose gas of 6,000 to 470,000 atoms of $^{133}$Cs at a temperature of $2–232$ nK in a three-dimensional harmonic trap. We tune the temperature and atom number by changing the trap depth at the end of the evaporation process. The harmonic trap frequencies range from $(\omega_x, \omega_y, \omega_z) = 2 \pi \times (24, 13, 74)$ to $2 \pi \times (36, 15, 91)$ Hz. The atoms are polarized into the hyperfine ground state $|F = 3, m_F = 3\rangle$, where $F$ and $m_F$ are the quantum numbers for the total spin and its projection along the magnetic-field direction, respectively. The narrow $g$-wave Feshbach resonance couples Cs atoms into Cs$_2$ molecules at $B = 19.849(1)$ G. The quantum numbers for the sum of the spins of two individual atoms and the orbital angular momentum of a molecule, respectively, are $m_l$ and $m_f$ along the magnetic-field direction, respectively.

To induce the molecule formation dynamics, we quench the magnetic-field close to the resonance position $B_0$ at 19.5 G where the samples are prepared. After holding for variable times, we switch the field back to either 19.5 G or 17.17 G to decouple atoms and molecules. We can image the remaining atoms at this field by absorption imaging. We also can wait for the remaining atoms to fly away after a resonant light pulse and image the molecules by jumping the field up to 20.4 G to dissociate them into atoms and then image the atoms from the dissociation. For the atom loss measurements shown in Fig. 3c, BECs with ~40,000 atoms are transferred from the harmonic trap to a 2D flat-bottomed optical potential before we quench the field to different values near the resonance. For the rest of the data shown in Figs. 2–4, we start from atomic samples in the three-dimensional harmonic dipole trap.

To measure the temperature of atoms or molecules (for example, Fig. 3b), we release them into a horizontally isotropic harmonic trap for a quarter of the trap period, which converts the particle distribution from real space to momentum space. We extract the temperature $T$ by fitting the momentum distribution with the condensate around zero momentum excluding the Gaussian function $n(k_f) = m(0) \exp(-h^2 k_f^2 / (2m k_B T))$, where $k_f$ is the radial wavenumber and $k_B$ is the Boltzmann constant.

Determination of Feshbach resonance position and width
To determine the position of the narrow $g$-wave Feshbach resonance in our system, we perform measurements of molecular binding energy at different offset magnetic fields using magnetic-field modulation spectroscopy and find the field value where the binding energy reaches zero.

We start with atomic BECs at 23 nK prepared at 19.5 G. Then, we quench the field to an offset value $B_{\text{bg}}$, near the resonance and simultaneously modulate the field sinusoidally with an amplitude $B_{\text{bg}} = 5$ mG for 5 ms. We scan the modulation frequency and measure the spectrum of the remaining atom number. From the atom loss peak of the spectrum due to the conversion from atoms into molecules, we extract the resonant frequency that corresponds to the molecular energy at an offset magnetic field $B_{\text{bg}}$ near the $g$-wave Feshbach resonance (Extended Data Fig. 1). We have confirmed that the resulting peak position of atom loss is not sensitive to the modulation amplitude and modulation time. A linear fit to the data in Extended Data Fig. 1 gives the resonance position $B_0 = 19.849(1)$ G where the molecular energy goes to zero. The slope of the linear fit gives the magnetic-moment difference between two bare atoms and one bare molecule as $\delta \mu = \hbar \times 0.76(3)$ MHz G$^{-1}$, which is consistent with another work. We emphasize that for the narrow resonance we are using, the molecular energy quadratically approaches zero only within a small fraction of the resonance width. Our linear fit to the molecular energy data underestimates the resonance position by ~0.3 mG based on our calculation using the resonance width from the following scattering length measurements.


determination of the absolute magnetic field is less than 20 mG. Throughout this work, we perform the magnetic-field calibration based on the same procedure to ensure a constant systematic error.

Next, we measure the $s$-wave scattering length near resonance to obtain the resonance width. Here the scattering length is inferred from the expansion of a quasi-2D BEC prepared with trap frequencies $(\omega_x, \omega_y, \omega_z) = 2 \pi \times (11, 13, 895)$ Hz. During the expansion, the mean-field interaction energy is converted into kinetic energy. We first prepare the BEC at an initial magnetic field $B = 20.481$ or 19.498 G where the scattering is $a$. The column density distribution of atoms in the Thomas–Fermi regime is 

$$n(x, y) = \frac{\mu - \frac{1}{2} m \omega_x^2 x^2 - \frac{1}{2} m \omega_y^2 y^2}{g_s}.$$  

where $g_s = (h^2 / m) \sqrt{\delta \mu / \mu}$ is the coupling strength, $\mu = \sqrt{\mu / \mu}$ is the harmonic oscillator length in the tightly confined $x$ direction. $T$ is the chemical potential determined by $B_0$, the total atom number $N$ and the initial trap frequencies $\omega_x$ and $\omega_y$ in the horizontal plane. Then, we quench the magnetic field to a different value $B_0$ where the scattering length is $a_0$ and simultaneously switch off the harmonic trap in the horizontal plane. According to another work, the dynamics of a BEC after release follow a simple dilation with scaling parameters $\lambda_x(t)$ and $\lambda_y(t)$, which determine the density distribution at time $t$ as 

$$n(x, y, t) = \frac{\mu - \frac{1}{2} m \omega_x^2 x^2 - \frac{1}{2} m \omega_y^2 y^2}{g_s \lambda_x(t) \lambda_y(t)}.$$  

where the scaling parameters evolve according to 

$$\lambda_x(t) = \frac{a_0}{\lambda_y(\omega_0/\omega_s(t))}$$  

$$\lambda_y(t) = \frac{a_0}{\lambda_x(\omega_0/\omega_s(t))}.$$  

We scan the magnetic field and measure the Thomas–Fermi radii $B_j = \sqrt{2 \mu j^2 / m a_j^2}$ where $j = x, y$ after 20 ms expansion. Eventually, we extract $a_0$ based on its one-to-one correspondence to the Thomas–Fermi radii according to equation (6). The results are summarized in Extended Data Fig. 2 and we fit the scattering length data using the formula 

$$a(B) = a_{bg} \left[1 + \eta(B - B_0) \left(1 - \frac{\Delta B}{B - B_0}\right)\right].$$

where we obtain the resonance width $\Delta B = 8.3(5)$ mG, the resonance position $B_0 = 19.861(1)$ G, the background scattering length on resonance $a_{bg} = 163(1)$a$_0$ and the slope of the background scattering length $\eta = 0.31(2)$ G$^{-1}$. The background scattering length $a_{bg}$ and slope $\eta$ are consistent with ref. 36 and the resonance width $\Delta B$ is consistent with ref. 37, where a different method is used. The fitted resonance position deviates from that in the binding energy measurement by ~10 mG, which we attribute to the heating of atoms near the resonance. The binding energy measurement, however, suffers less from the heating issue. Throughout this paper, we adopt the resonance position $B_0 = 19.849(1)$ G from the binding energy measurement.

Extraction of molecule oscillation frequency, atom loss rate and molecule formation rate
We use the following function to fit the data in the equilibrium phase at $1 < t < 3$ ms (Fig. 3) for the extraction of molecule oscillation frequencies:

$$N_m(t) = N_m(0) - \gamma t + \Delta N_m e^{-\gamma t} \sin(\omega t + \phi).$$

The systematic error of our calibration of the absolute magnetic field is less than 20 mG. Throughout this work, we perform the magnetic-field calibration based on the same procedure to ensure a constant systematic error.
where the fitting parameters are the molecule number $N_m(0)$ extrapolated to time $t = 0$, decay rates $\gamma_1$ and $\gamma_2$, oscillation amplitude $\Delta N_m$, oscillation frequency $\omega$ and phase $\phi$. Here the two decay rates $\gamma_1$ and $\gamma_2$ characterize the decay of molecule number and damping of molecule oscillation amplitude, which are generally different.

For the data shown in Fig. 4, we fit the data at $0.3 < t < 3.0$ ms using the function

$$N_m(t) = e^{-\gamma t}[N_m(0) + \Delta N_m \sin(\omega t + \phi)],$$

where we find that the single decay rate $\gamma_1$ is enough to describe the data very well. For each fit, we subtract a delay time of 0.15 ms from time $t$ due to the finite speed of our magnetic-field switch.

To prevent the fits from getting stuck in a local optimum, we vary the initial guess of frequency $\omega$ for the fits and use the result that has the minimum root mean square error.

For the atom loss rate measurement shown in Fig. 3c, we present example time traces of the averaged atomic density in the 2D flat-bottomed trap (Extended Data Fig. 3). Far from the resonance (Extended Data Fig. 3a,b), the atomic density decays slowly and we fit the data using

$$n_a(t) = n_a(0)e^{-\gamma t},$$

where $n_a(0)$ is the initial atomic density and $\gamma$ is the atom loss rate. The fit is applied to the data above half of the initial density.

Below and near the resonance (Extended Data Fig. 3c), we find that the density first decays rapidly and then settles around some equilibrium value before a slow decay kicks in at a timescale longer than 3 ms. In this case, we use the following fit function:

$$n_a(t) = n_a(0)\{\theta(t_0 - t) + [(1 - s)e^{-\gamma_1(t-t_0)} + s]e^{-\gamma_2(t-t_0)}\},$$

where $t_0$ is the time when the decay begins and $s$ represents the fractional density that the system settles to after the initial fast decay. On the other hand, above and near the resonance (Extended Data Fig. 3d), the data are fit well by a single exponential decay (equation (10)). Here we also provide the molecule formation rate measured near the resonance, complementary to the atom loss rate measurements shown in Fig. 3c (Extended Data Fig. 4).

**Data availability**

Source data are provided with this paper. All other data that support the plots within this paper are available from the corresponding author upon reasonable request.

**Code availability**

The codes for the analysis of data shown in this paper are available from the corresponding author upon reasonable request.

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**Author contributions**

Z.Z. and S.N. performed the experiments and analysed the data. Z.Z. built the theoretical model. K.-K.Y. contributed to the discussion of the results. C.C. supervised the work. All authors contributed to the writing of the paper.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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Extended Data Fig. 1 | Bound state energy diagram for cesium atoms in the hyperfine ground state \( |F = 3, m_F = 3 \rangle \) and molecular energy measurement near the g-wave Feshbach resonance around 20 G using modulation spectroscopy. **a.** Energy diagram for Cs\(_2\) molecular states close to the atomic scattering continuum adapted from Fig. 22 in Ref. 19. **b.** Molecular energy \( \varepsilon_m \) obtained from modulation spectroscopy at different offset magnetic fields. The solid line is a linear fit which reaches 0 at \( B_0 = 19.849(1) \) G.
Extended Data Fig. 2 | Scattering length measurement near the narrow g-wave Feshbach resonance by time-of-flight.  

**a**, Atomic density distributions after 20 ms time-of-flight at different magnetic fields near the Feshbach resonance. The images with $B < 19.865 \text{ G}$ ($B > 19.865 \text{ G}$) come from initial BECs prepared below (above) the Feshbach resonance.  

**b**, Scattering length extracted from the Thomas-Fermi radii in the time-of-flight images, see text. The circular (diamond) data points come from initial BECs prepared below (above) the Feshbach resonance. The solid line is a fit to the data excluding the points at $19.858 \text{ G} < B < 19.909 \text{ G}$ based on Eq. (7), from which we obtain the resonance width $\Delta B = 8.3(5) \text{ mG}$. The points at $19.855 \text{ G} < B < 19.909 \text{ G}$ are excluded because of the heating effect near the resonance.  

**c**, Total atom number extracted from the time-of-flight images.
Extended Data Fig. 3 | Examples of atomic density evolution in a 2D flat-bottomed optical potential for the data presented in Fig. 3c. For data below the resonance, BECs are initially prepared at 19.5 G and magnetic field is quenched to values between 0.05 and 1 G (panel a) and between 5 and 50 mG (panel c) below the resonance. Relaxation and equilibration phases are marked with different background colors in panel c. For data above the resonance, BECs are initially prepared at 20.4 G and magnetic field is quenched to values between 0.1 and 1 G (panel b) and between 10 and 50 mG (panel d) above the resonance. Solid lines are fits for extracting the atom loss rates, see text.
Extended Data Fig. 4 | Molecule formation rate near the resonance complementary to the atom loss rate measurements in Fig. 3c. Solid (empty) circles represent samples prepared below (above) the resonance.