**Materials and Methods**

**Magnetic levitation field**

For the compensation of gravity by a magnetic gradient force, we use a quadrupole field symmetric with respect to a vertical axis ($z$-axis) through the center of the atomic or molecular cloud. The field is produced by two pairs of coils, which provide independent control of the vertical field gradient $B'$ and the bias field $B_0$. While the inner anti-Helmholtz pair provides the gradient $B'$, the outer Helmholtz pair generates the bias field $B_0$. The field experienced by a particle at a vertical position $z$ on the symmetry axis is given by $B_0 + B'z$.

![Fig. S1](image)

*Fig. S1.* Illustration of the magnetic levitation field. (A) Two pairs of coils facilitate independent control of the magnetic bias field $B_0$ (blue) and the vertical field gradient $B'$ (red). (B) The contour lines of equal magnetic fields are centered around the magnetic zero, which is located at a distance of $B_0/B'$ below the ultracold cloud. The curvature leads to a weak outward directed antitrapping force, depending on the distance $x$ from the symmetry axis.

The levitation of a high-field seeking particle can be understood by looking at the repulsive effect of the magnetic quadrupole which forms an axially symmetric antitrap with its center located below the levitated particles. Above the magnetic-field zero, the increasing magnetic field pushes the particles upwards. On the symmetry axis, gravity can be exactly cancelled in this way. Off axis, however, the magnetic force is not directed exactly in vertical direction. To
first order in the distance $x$ from the symmetry axis, the levitation condition is not affected, but a weak outward directed force occurs in the horizontal direction with anti-trapping character. This horizontal force can be written as

$$F_x = m\alpha^2 x.$$ 

For exact levitation, the characteristic rate is given by

$$\alpha = \frac{g}{2\sqrt{\frac{m}{\mu B_0}}},$$

where $m$ and $\mu$ are the particle’s mass and magnetic moment, respectively, and $g$ denotes the gravitational acceleration.

**Experimental timing and magnetic-field ramps**

The starting point of the ramp is above the resonance at 19.88 G. With a ramp-speed of 48 G/s the field is swept over the resonance to a final field value of 19.74 G. Immediately after the end of the ramp we switch off the optical trap and ramp the gradient field within 0.5 ms to about 50 G/cm to cancel the gravitational force for the molecules. Simultaneously the magnetic field is lowered in 2 ms to a fixed hold field of 17 G where the molecules are allowed to expand under the influence of the gradient field.

In the time between the production of the molecules and the end of the gradient field ramp, the molecules are initially accelerated with 0.38 g. This leads to a vertical center-of-mass velocity of typically a few mm/s, depending on the exact timing of the ramps.

For detection we dissociate the molecules to atoms by ramping back over the resonance with a ramp-speed of 1000 G/s. The ramp takes 4 ms and reaches a final value of 21 G, well above the resonance. For ramp-speeds ranging from 300 G/s up to 1100 G/s, we did not find any influence on the dissociation process. The instant of time of the dissociation depends on the local magnetic field and thus on the vertical position of the molecular cloud in the gradient field. With a finite vertical velocity the position varies with expansion time. By switching off the gradient field at the start of the ramp ($t_1$), we ensure that the time of dissociation is fixed. Nevertheless the exact time ($t_{\text{diss}}$) is not exactly known due to an unknown offset in the magnetic field caused by switching the gradient field. The expansion in the time between $t_1$ and $t_{\text{img}}$ ($\Delta t = 5.5$ ms) takes place without the influence of the curvature of the gradient field.

By varying the time between $t_{\text{diss}}$ and $t_{\text{img}}$ we can characterize the dynamics of the reconverted atoms. We observe a fast expansion with an energy of about $\frac{1}{2}k_B \times 1\mu\text{K}$. This, together
with the uncertainty in $t_{\text{diss}}$, leads to a fixed additional broadening ($\sigma_{\text{res}}$) of the cloud. We switch off the magnetic bias field 1.5 ms before the imaging. The switching speed is much too fast to convert atoms into molecules when the Feshbach resonance is crossed again. At $t_{\text{img}}$ the cloud is imaged using a 100 $\mu$s resonant absorption pulse.

The measured center-of-mass velocity suggests that the production of the molecules takes place in the center of the initial creation ramp, with an uncertainty of 1 ms, and this defines the point where $t = 0$. Hence, the total time of expansion is measured from $t = 0$ until the time of imaging $t_{\text{img}}$.

**Measurement of the magnetic moment**

The compensation of gravity by the magnetic gradient field facilitates a precise measurement of the magnetic moment of the molecular state. The basic idea is to derive this molecular moment by comparing the field gradient needed for levitation of the molecules with the one required for the atoms. The known magnetic moment of the atoms serves as a reference.

Figure S3 shows the vertical center-of-mass motion of the molecular cloud in a magnetic field of 17 G (at $z = 0$) with a gradient of 49.7 G/cm, close to the exact levitation condition. A
Fig. S3. Measurement of the molecular magnetic moment. The vertical center-of-mass position of the molecular cloud at a bias field of 17 G (at $z = 0$) is shown as a function of time together with a parabolic fit (solid line). The applied magnetic gradient field of 49.7 G/cm is slightly below the exact levitation condition so that a residual downward-directed acceleration is observed.

A parabolic fit to the data yields a residual acceleration of $0.11(5)$ m/s$^2$. Taking this small correction into account we calculate a gradient of $50.2(3)$ G/cm for exact levitation of the molecules. The magnetic moment of the molecular state is then determined to $0.930(5)$ $\mu_B$.

The fit to the data of Fig. S3 also yields an initial center-of-mass velocity of the molecular cloud of $4(1)$ mm/s. This velocity results from the downward acceleration before the field gradient increases to the molecule levitation value.

**Horizontal expansion in the levitation field**

According to the equation of motion $F_x = m\alpha^2 x$ for the horizontal degrees of freedom, a particle released at $t = 0$ in the levitation field with an initial horizontal position $x_0$ and an initial horizontal velocity $v_0$ follows

$$
\begin{align*}
x(t) & = x_0 \cosh(\alpha t) + \alpha^{-1} v_0 \sinh(\alpha t), \\
v(t) & = \alpha x_0 \sinh(\alpha t) + v_0 \cosh(\alpha t).
\end{align*}
$$

In our experiment, the levitation field is turned off a time $\Delta t$ before the image is taken at $t_{img}$. In this short final time interval, beginning at $t_1 = t_{img} - \Delta t$ and ending at $t_{img}$, a free motion in the field of gravity takes place. At the time of detection, the horizontal position of the
particle is

\[ x(t_{\text{img}}) = x(t_1) + v(t_1)\Delta t = x_0 \left[ \cosh(\alpha t_1) + \alpha \Delta t \sinh(\alpha t_1) \right] + v_0 \left[ \alpha^{-1} \sinh(\alpha t_1) + \Delta t \cosh(\alpha t_1) \right] \]

For an ensemble of particles with statistical initial distributions of \(x_0\) and \(v_0\) we introduce \(s_x\) and \(u_x\) for the horizontal position spread and velocity spread (root-mean-square values). For the horizontal size of the cloud we then obtain the rms value

\[ \sigma_x = \sqrt{s_x^2 \left[ \cosh(\alpha t_1) + \alpha \Delta t \sinh(\alpha t_1) \right]^2 + u_x^2 \left[ \alpha^{-1} \sinh(\alpha t_1) + \Delta t \cosh(\alpha t_1) \right]^2}. \]

**Expansion data: fit procedure and error analysis**

In order to extract the relevant parameters from the observed expansion of the molecular cloud (Fig. 4) we proceed in the following way.

**Vertical expansion.** For the vertical size of the expanding cloud we use the fit function

\[ \sigma_{z, \text{img}} = \sqrt{\sigma_{\text{res}}^2 + s_z^2 + u_z^2 t_{\text{img}}^2}. \]

Here two free parameters are used: \(u_z\) denotes the initial rms velocity spread (predominantly resulting from the velocity dispersion), and \(\sigma_{\text{res}}\) is the spatial resolution limit that results from the dissociation energy in the detection phase. The fixed parameter \(s_z = 3.7 \, \mu m\) is the initial vertical rms position spread that we calculate from the Thomas-Fermi distribution of the BEC.

For the velocity spread the fit yields \(u_z = 1.12(5) \, \text{mm/s}\), corresponding to a mean kinetic energy of \(E_z = \frac{1}{2} k_B \times 40(3) \, \text{nK}\). For the resolution parameter we obtain \(\sigma_{\text{res}} = 11(3) \, \mu m\). The given errors denote the one-standard-deviation (one sigma) values as resulting from the fit.

**Horizontal expansion.** For the horizontal expansion in the levitation field, we use the above expression for the cloud size \(\sigma_x\) together with the resolution parameter \(\sigma_{\text{res}}\) combined to the fit function

\[ \sigma_{x, \text{img}} = \sqrt{\sigma_{\text{res}}^2 + \sigma_x^2} = \sqrt{\sigma_{\text{res}}^2 + s_x^2 \left[ \cosh(\alpha t_1) + \alpha \Delta t \sinh(\alpha t_1) \right]^2 + u_x^2 \left[ \alpha^{-1} \sinh(\alpha t_1) + \Delta t \cosh(\alpha t_1) \right]^2}. \]
where \( t_1 = t_{\text{img}} - \Delta t \). The two free parameters of the fit are the initial horizontal cloud size \( s_x \) and an initial velocity spread \( u_x \). The parameter \( \alpha = 27.2 \text{s}^{-1} \) is calculated for the effective horizontal curvature of the levitation field. The resolution parameter \( \sigma_{\text{res}} = 11(3) \mu\text{m} \) is taken from the above fit to the vertical expansion and varied within its error range. From this fit procedure we obtain the initial horizontal cloud size \( s_x = 20(2) \mu\text{m} \). For the initial rms velocity spread the fit yields \( u_x = 0.22(16) \text{mm/s} \), which corresponds to a mean initial kinetic energy spread of \( E_x = \frac{1}{2} k_B \times (40 \pm 3 \pm 2) \text{nK} \) vertically, \( E_x = \frac{1}{2} k_B \times (2 \pm 2 \pm 3) \text{nK} \) horizontally, where the two given error numbers denote the one-standard-deviation statistical error (first error) and the systematic error (second error).

**Expansion energies and total error budget.** In addition to the given statistical errors, systematic errors stem from the limited knowledge of the experimental parameters used in the above fits. A more detailed analysis shows that all systematic errors are far below the statistical errors with the exception of the origin of time, i.e. the instant at which the Feshbach conversion produces the molecules. We have therefore repeated the above fits with a time origin varied in the maximum possible error range of \( \pm 1 \text{ms} \). The corresponding results indicate systematic deviations from the above fit results which can be taken as the systematic errors.

For the most interesting quantities, the initial kinetic-energy spreads of the expanding molecular cloud, we finally obtain

\[
E_z = \frac{1}{2} k_B \times (40 \pm 3 \pm 2) \text{nK} \quad \text{vertically,}
\]
\[
E_x = \frac{1}{2} k_B \times (2 \pm 2 \pm 3) \text{nK} \quad \text{horizontally,}
\]