In developing micro- and nanodevices, stiction between their parts, that is, static friction preventing surfaces in contact from moving, is a well-known problem. It is caused by the finite-temperature analogue of the quantum electrodynamic Casimir–Lifshitz forces, which are normally attractive. Repulsive Casimir–Lifshitz forces have been realized experimentally, but their reliance on specialized materials severely limits their applicability and prevents their dynamic control. Here we demonstrate that repulsive critical Casimir forces, which emerge in a critical binary liquid mixture upon approaching the critical temperature, can be used to counteract stiction due to Casimir–Lifshitz forces and actively control microscopic and nanoscopic objects with nanometre precision. Our experiment is conducted on a microscopic gold flake suspended above a flat gold-coated substrate immersed in a critical binary liquid mixture. This may stimulate the development of micro- and nanodevices by preventing stiction as well as by providing active control and precise tunability of the forces acting between their constituent parts.
casimir–lifshitz forces (dashed orange line, eq. (5)) and the critical casimir-hydrophilic flake above a hydrophilic (blue layer, inset) surface as a function of its height b. since the boundary conditions are symmetric (-,-), both the casimir–lifshitz forces (dashed orange line, eq. (5)) and the critical casimir forces (dashed blue line, eq. (2)) at ΔT = Tc = −0.1 K, where T is the solution temperature and Tc is the critical temperature of the critical binary mixture) are attractive. the total force (black line) including also a repulsive electrostatic force component (dashed green line) vanishes at h = 80 nm. c, forces on a hydrophilic (-) flake above a hydrophobic (+) surface (red layer, top inset) as a function of h. here, the antisymmetric (-,+) boundary conditions induce a repulsive critical casimir force (dashed red line, eq. (2) at ΔT = −0.1 K), while the casimir–lifshitz force (dashed orange line, eq. (5)) remains attractive. accordingly, the total force vanishes at much larger h = 210 nm (bottom inset shows a zoom-in view of this region). the presence of repulsive critical casimir forces greatly raises the equilibrium height of the flake above the surface. the forces shown in b and c are calculated for a 34 nm-thick 1.520 nm-wide gold flake suspended in water-2.6-lutidine above a 40 nm-thick gold layer.

surfaces, leading to the formation of Fabry–Pérot cavities when multiple particles assemble on top of each other53. despite these advances in the manipulation of nanoparticles, these systems are inherently static and lack any tunable parameters to allow dynamic control. although a protocol to dynamically control the casimir–lifshitz force was proposed theoretically in ref. 54 by varying the strength and orientation of a magnetic field, its experimental realization is still challenging.

Critical casimir forces are the thermodynamic analogue of quantum electrodynamical casimir forces and were first theoretically predicted by Fisher and de gennes in 197855. they arise, for example, between objects immersed in a binary liquid mixture kept near a second-order phase transition, owing to the confinement of the thermal fluctuations of the local concentration of one of the components of the binary mixture (which is the order parameter of the transition)56. In soft matter, thermal fluctuations typically occur on molecular length scales (subnanometre) and, accordingly, the effects they produce are generally negligible at larger separations. however, upon approaching the critical point of a second-order phase transition, the fluctuations become correlated on much longer length scales (up to several microns) and can thus substantially affect the behaviour of microscopic systems57. importantly, critical casimir forces can be either attractive or repulsive depending on the surface chemistry of the objects involved58–60. while the critical casimir forces between surfaces with similar adsorption preferences (either hydrophilic or hydrophobic) are attractive, they become repulsive when these preferences are opposite.

Fig. 1 | Casimir–Lifshitz and critical Casimir forces between parallel plates. a, schematic (not to scale) of a hydrophilic (-) gold flake hovering at an equilibrium height h above a glass surface coated with a gold layer and treated with sams to control the preferential surface adsorption. b, forces acting on a hydrophilic flake above a hydrophilic (blue layer, inset) surface as a function of its height. since the boundary conditions are symmetric (-,-), both the casimir–lifshitz forces (dashed orange line, eq. (5)) and the critical casimir forces (dashed blue line, eq. (2)) at ΔT = Tc = −0.1 K, where T is the solution temperature and Tc is the critical temperature of the critical binary mixture) are attractive. the total force (black line) including also a repulsive electrostatic force component (dashed green line) vanishes at h = 80 nm. c, forces on a hydrophilic (-) flake above a hydrophobic (+) surface (red layer, top inset) as a function of h. here, the antisymmetric (-,+) boundary conditions induce a repulsive critical casimir force (dashed red line, eq. (2) at ΔT = −0.1 K), while the casimir–lifshitz force (dashed orange line, eq. (5)) remains attractive. accordingly, the total force vanishes at much larger h = 210 nm (bottom inset shows a zoom-in view of this region). the presence of repulsive critical casimir forces greatly raises the equilibrium height of the flake above the surface. the forces shown in b and c are calculated for a 34 nm-thick 1.520 nm-wide gold flake suspended in water-2.6-lutidine above a 40 nm-thick gold layer.

The first direct experimental measurement of critical casimir forces was achieved only in 200859: using total internal reflection microscopy, femtonewton forces were measured on a single spherical microscopic particle suspended above a planar surface in a critical water–ludidine liquid mixture. since then, these forces have been investigated under various conditions, for example, by varying the properties of the surfaces involved, demonstrating the occurrence of both attractive and repulsive forces59–64 as well as the existence of many-body effects46 and the possibility of levitating spherical colloids over patterned substrates solely by critical casimir forces45. the subtle interplay between these forces and the electrostatic interaction in the presence of salt45–48 or the van der waals force in nonpolar fluids47 have also been investigated. studies of the phase behaviour of colloidal dispersions in a critical mixture50,59 have indicated that critical casimir forces are viable candidates to control the self-assembly of micro- and nanostuctures46,48 and quantum dots51. in addition, fluctuation-induced effects similar to critical casimir forces have been investigated, for example, at the percolation transition of a chemical sol52, in the presence of temperature gradients46, in granular fluids60 and in active matter systems60,61.

Here we demonstrate experimentally that repulsive critical casimir forces can be used to compensate for attractive casimir–lifshitz forces. in particular, we study a microscopic gold flake diffusing above a gold surface immersed in a critical binary liquid mixture. we control the magnitude of these forces by varying the temperature of the mixture and the attractive/repulsive character of the critical casimir forces by chemical functionalization of the gold flake and surface, obtaining repulsive forces for opposite surface functionalizations. far from the critical temperature, we observe stiction between the flake and the surface due to casimir–lifshitz forces. however, as we approach the critical temperature, we observe the emergence of repulsive critical casimir forces that are sufficiently strong to prevent stiction and to release the flake from the surface, above which it then levitates. in addition, we demonstrate that this behaviour is reversible and can be employed to actively control the flake position above a structured surface.

results

working principle

We first describe a simple model to clarify how repulsive critical casimir forces can be employed to counteract attractive casimir–lifshitz forces, by estimating theoretically the orders of magnitude of the forces involved. consider two flat parallel plates (fig. 1a): one is a thin gold flake (length a = 1.520 nm, thickness b = 34 nm) suspended in a critical binary liquid mixture above a gold-coated (thickness 40 nm) glass substrate, which is the second plate. the first plate hovers at a height h above the surface because of the interplay of three forces (see details in methods): the casimir–lifshitz force, which, in this case, is attractive; the critical casimir force, whose character and strength can be varied by modifying the surface functionalization and tuning the solvent
temperature; and the electrostatic force, which provides a short-range repulsion between the flake and the substrate. The gravitational force acting on the flake is negligible compared with the other forces acting on the flake in close proximity to the substrate.

The attractive Casimir–Lifshitz force between two plates depends on $h$ as

$$F_{\text{CL}}(h) = -\frac{dG_{\text{CL}}}{dh} S,$$

where $G_{\text{CL}}$ is the Casimir–Lifshitz free energy per unit area of the system and $S$ is the area of the flake (ref. 46 and Methods). In this case, the force depends on the thicknesses of the flake and of the gold layer deposited on the glass surface, as well as on the dielectric properties of the glass and of the solvent (Methods and Extended Data Fig. 1). The resulting Casimir–Lifshitz forces are plotted as dashed orange lines in Fig. 1b,c for two opposite surface treatments of the gold layer.

The critical Casimir forces are induced by the confinement of the critical mixture between the flake and the substrate. Their strength $F_{\text{crit}}$ depends on the adsorption preference of the surfaces involved and on the difference $\Delta T = T - T_c$ between the actual temperature $T$ of the binary liquid mixture and its critical temperature $T_c$. In particular,

$$F_{\text{crit}}(h, \Delta T) = -\frac{k_B T_c}{h^3} \theta(h/\xi(\Delta T)) S,$$

where $\xi(\Delta T)$ is the correlation length of the fluctuations of the order parameter of the mixture depending on $\Delta T$ and $\theta(h/\xi(\Delta T))$ is a universal scaling function. For the sake of simplicity, in the following we omit the explicit dependence on $\Delta T$ of $\xi$.

Whether the critical Casimir forces are repulsive or attractive depends on the boundary conditions given by the surface preferential adsorption of one of the components of the mixture (indicated by “$-$” or “$+$”). We can control these boundary conditions by chemically functionalizing the surfaces with appropriate self-assembled monolayers (SAMs) (Methods). In the case of symmetric boundary conditions (−−) or (++) (that is, the flake and surface prefer the same component of the mixture), the critical Casimir forces are attractive (Fig. 1b, dashed blue line). For antisymmetric boundary conditions (−+) (that is, preference for different components), the critical Casimir forces are repulsive (Fig. 1c, dashed red line).

The electrostatic repulsive force $F_{\text{ES}}$ is due to the double of counterions in close proximity to the surface of the flake and the substrate. According to the Derjaguin–Landau–Verway–Overbeek (DLVO) theory (neglecting boundary effects), we have

$$F_{\text{ES}}(h) = P_0 e^{-h/\lambda_0} S,$$

where $\lambda_0$ is the Debye screening length of the critical mixture and $P_0$ is a parameter with the dimensions of pressure.

The theoretical predictions presented in Fig. 1b,c show that, in the configuration we consider at $\Delta T = -0.1 K$, the magnitude of the critical Casimir forces is expected to be larger than that of the Casimir–Lifshitz forces, so we can use the critical Casimir forces to tune the equilibrium position of the flake above the surface. In the case of asymmetric boundary conditions (−+, Fig. 1c), this provides an additional repulsion between the flake and surface, which can be used to prevent stiction.

**Experimental setup and analysis**

In the experiment, we consider a gold flake suspended in a near-critical water–2,6-lutidine mixture (lutidine critical concentration $c^*_L = 0.286$ mass fraction, lower critical temperature $T_L = 310$ K, that is, $34^\circ$C) above a gold-coated surface (Fig. 1a). The gold flakes are fabricated using a wet chemical synthesis method that produces single-crystalline gold flakes (diameter $d = 3 \pm 2 \mu m$, stabilized by cetrimonium bromide (CTAB) ligand molecules, with average thickness of $b = 34 \pm 10$ nm in aqueous solution and of various geometrical shapes (scanning electron microscopy image in Fig. 2a and details in Methods). These flakes are hydrophobic due to the formation of a CTAB bilayer on their surface. For this study, we select flakes with the shape of equilateral hexagons (Fig. 2b, inset) owing to their larger degree of symmetry.

The substrate is constituted by a glass slide on which a 35-nm-thick gold coating is deposited by sputtering. To control the surface preferential adsorption, the gold coating is chemically functionalized by a SAM of either hydrophilic or hydrophobic thiols by immersing the surface in a 1 mM thiol solution with different end groups (Methods). We tune the temperature of the sample via a two-stage feedback temperature controller with $\pm 20$ mK stability and thus control the strength of the critical Casimir forces.

As anticipated by our theoretical predictions (Fig. 1 and discussion above), a change in the magnitude or sign of the critical Casimir force acting on the flake alters its equilibrium height $h$ above the surface. However, these changes are on the order of a few tens of nanometres and, thus, difficult to measure directly. A more convenient approach consists in measuring the lateral diffusion of the flakes along the substrate, which depends sensitively on their height: the (lower) the flake is above the surface, the larger (smaller) its diffusion is.

Accordingly, we record a video of the flake's motion for a long period of time (~100 s, with a sample rate of 100 Hz) using a bright-field microscope (Extended Data Fig. 2). Using digital video microscopy, we reconstruct the projection of the flake's position on the $x$–$y$ plane, where $x$ and $y$ are the Cartesian coordinates along the surface of the substrate (Fig. 2b, solid line and Supplementary Video 1). The flake diffuses freely along the $x$–$y$ plane, while its motion along the $z$-direction (perpendicular to the substrate) is negligible. From the $x$–$y$ trajectory, we calculate the mean square displacement (MSD) at time $t$ in Fig. 2c, which, for a freely diffusing flake, grows as $\text{MSD}(t) = 4Dt$, where $D$ is the diffusion constant. $D$ can be obtained by fitting the measured MSD (Methods).

Since $D$ depends sensitively on $h$, its value can be used to infer the equilibrium position $h$ of the flake above the surface. We have calibrated the theoretical reference $D(h)$ (Fig. 2d, blue line) using hydrodynamic simulations of the flake (Methods), which permit a determination of the equilibrium height of the flake with nanometre precision (Fig. 2d, black dashed lines). In addition, we validated the indirect measurement of $h$ obtained as described above by alternatively measuring the Fabry–Pérot cavity modes (Supplementary Information Section II). In fact, a microcavity is formed between the flake and the gold substrate. We measure its reflectivity spectrum to determine the spatial variation of the cavity thickness using a spectrometer (Methods and Extended Data Fig. 3). The measured cavity modes in the reflection spectrum at different positions were analysed by the standard transfer matrix method.

We find that the flake is about $90 \pm 10$ nm away from the surface inside a critical water–2,6-lutidine mixture at $\Delta T = -0.5$ K, independently of the actual surface functionalization, and this value is in good agreement with our measurements of $h = 100$ nm using hydrodynamic simulations.

**Measurement of the Casimir–Lifshitz force**

Using the method described in the previous section, we can investigate the separate effects of the various forces on the flake. We start by determining the (attractive) Casimir–Lifshitz force at low temperature $\Delta T = -1$ K, at which the critical Casimir force is negligible (importantly, the Casimir–Lifshitz force and the electrostatic force are not substantially affected by a temperature change up to several kelvin, because they are expected to be smooth functions of the absolute temperature $T$, whose value changes by less than 1% in our experiment). The Casimir–Lifshitz force can then be determined by comparing the total force (which includes electrostatics) acting on the gold flake above the uncoated glass surface (where the Casimir–Lifshitz force is weak) and the force acting when the flake is above a gold-coated surface. In fact, we observe that the
flakes suspended above an uncoated glass surface hovers at \( h = 300 \) nm (Extended Data Fig. 4), while its equilibrium height reduces to \( h = 100 \) nm, when the flake is floating above the gold-coated surface. This fact is largely independent of the gold functionalization and, therefore, of the adsorption properties of the surface (as seen from the values of \( h \) at low temperature, \( \Delta T = -1 \) K, in Fig. 3b,c). This reduction of the value of \( h \) is the result of the presence of attractive Casimir–Lifshitz forces between the gold flake and the gold surface. Further evidence of the nature of this force is provided by the quality of the fit of the experimental data with the theoretical model (Extended Data Fig. 5). In particular, we compare the inferred heights with the theoretical average height of the flake floating above the gold-coated and uncoated silica surface. This fit allows us to infer the parameters of the electrostatic interaction in the two cases, which are important for the theoretical interpretation of the experimental results provided in the following two subsections. The details of the fitting procedure are given in Methods.

Interplay between critical Casimir and Casimir–Lifshitz forces

As seen in Fig. 1c, repulsive critical Casimir forces are theoretically expected to be able to overcome Casimir–Lifshitz forces in magnitude as the solvent’s temperature approaches \( T_c \). In this section, we provide experimental evidence for this fact.

In Fig. 3a–c, we consider a hexagonal hydrophilic flake with \( a = 700 \) nm (Fig. 2b) above a hydrophilic surface, realizing symmetric (−, +) boundary conditions. As the temperature of the solution is increased towards \( T_c \), the flake’s diffusion constant \( D \) decreases from \(-0.09 \mu m^2 s^{-1} \) to \(-0.07 \mu m^2 s^{-1} \) (blue squares in Fig. 3a), indicating that the flake’s distance from the substrate decreases from \( 108 \pm 7 \) nm to \( 68 \pm 5 \) nm (Fig. 3b, blue squares). This decrease is primarily due to an increasingly stronger critical Casimir force (Fig. 3c, blue symbols), but also to an increase of the Casimir–Lifshitz force (Fig. 3c, orange crosses) as the equilibrium distance \( h_{eq} \) of the flake decreases. The experimental results agree well with theoretical predictions for the Casimir–Lifshitz forces (Fig. 3c, solid orange line), the critical Casimir forces (Fig. 3c, solid blue line), the electrostatic force (Fig. 3c, solid green line) and the resulting height of the flake (Fig. 3b, solid blue line) (Methods). We emphasize that the magnitude of the critical Casimir forces acting on the flake becomes larger than that of the Casimir–Lifshitz forces when the temperature is sufficiently close to \( T_c \) (that is, \( \Delta T > -0.3 \) K) (Fig. 3c).

In Fig. 3d–f, we consider the case of a hexagonal hydrophilic flake with \( a = 840 \) nm above a hydrophobic surface realizing antisymmetric (+, +) boundary conditions. Interestingly, while the sizes of the two flakes are different, their equilibrium heights \( h_{eq} \) are similar, confirming that the forces acting per unit area are similar and not affected by the critical Casimir contribution, which would have opposite signs for the two flakes. In this case, however, as the temperature approaches \( T_c \), the flake’s diffusion constant \( D \) increases (Fig. 3d, red circles) from \(-0.067 \mu m^2 s^{-1} \) to \(-0.15 \mu m^2 s^{-1} \), indicating that its equilibrium height \( h_{eq} \) above the surface increases (Fig. 3e, red circles). In particular, \( h_{eq} \) changes from \( 96 \pm 10 \) nm at \( \Delta T = -0.9 \) K to \( 196 \pm 47 \) nm at \( \Delta T = -0.1 \) K. This increase is primarily due to the emergence of repulsive critical Casimir forces (Fig. 3f, red circles), combined with the decrease of the magnitude of the Casimir–Lifshitz forces (Fig. 3f, orange circles) as a consequence of the larger distance between the flake and the substrate. These experimental results fit well with the theoretical predictions (Fig. 3f, solid lines) (Methods). Also for this...
constant hydrophilic surface (symmetric (−, −) boundary conditions) decreases as the temperature $T$ approaches the critical temperature $T_c$, which is indicated as shaded areas for the confidence levels of 68%, 86% and 95%. In e and f, only the 68% confidence level error range is shown. The dashed lines in b and e refer to the equilibrium height of particle far away from criticality.

The electrostatic repulsion in the two cases with different boundary conditions is found to be essentially the same by the fitting procedure (Methods). Thus, this experiment demonstrates that the repulsive critical Casimir forces, as the only tunable repulsive force in the system, can effectively overcome the Casimir–Lifshitz attraction upon approaching the critical temperature $T_c$. Note that, in this case, the presence of the electrostatic force is not necessary to prevent stiction, at least close to the critical point. On the other hand, the electrostatic repulsion may be fundamental to avoid stiction when far from criticality or in the presence of attractive critical Casimir forces.

### Spatio-temporal control

In the following, we show experimentally how a particle confined to a metallic surface by Casimir–Lifshitz attraction can be lifted away from the substrate by the repulsive critical Casimir force and, therefore, start diffusing freely. For this simple, proof-of-principle experiment we fabricated gold stripes (width 3 μm) separated by gaps of glass of the same width (Fig. 4a). We treated the glass stripes and the gold stripes with SAMs having opposite wetting properties to make them hydrophilic and hydrophobic, respectively. A hydrophilic gold flake ($a = 1450$ nm) is floating above this configuration. The size of the flake is indicated in Fig. 4a and is comparable to the stripe width. Accordingly, symmetric boundary conditions (−, −) are realized when the flake is entirely above the glass stripes, while the boundary conditions are antisymmetric (+, −) when it is entirely above the gold stripes. To study the interplay between the Casimir–Lifshitz forces and the critical Casimir forces, the temperature of the mixture is modulated periodically in time (Fig. 4a, inset). At temperatures far below $T_c$ that is, with $\Delta T = -1.5$ K, the flake is essentially confined on the gold stripes by the Casimir–Lifshitz and electrostatic forces, respectively. Data presented as mean values. The error bars on the experimental points in b and e are the s.d. from three measurements. The error range for the theoretical lines in b and e is indicated as shaded areas for the confidence levels of 68%, 86% and 95%. In e and f, only the 68% confidence level error range is shown. The dashed lines in b and e refer to the equilibrium height of particle far away from criticality.
that transitions occur shortly after reaching $T_c$, which is appreciably below $T_c$ (white trajectories) so that the Casimir–Lifshitz force dominates and confines the flake over the gold stripes (dark stripes in background images) (b,d,f,h) and when $T = T_c$ (black trajectories) so that the critical Casimir force emerges, repelling the flake from the gold stripes to the glass regions (light stripes in background images) (c,e,g,i,j). Full trajectory of the flake under periodic heating and cooling of the sample, showing that transitions occur shortly after reaching $T = T_c$ (Supplementary Video S2). Its starting point is marked by the particle image. Scale bars 6 μm.

In the theoretical model describing a flake suspended on a substrate coated with gold stripes, the flake is subject to a potential $U_{exp}(\Delta T; x, h)$ (Extended Data Fig. 6a–b), which is a periodic function of the coordinate $x$ of the centre of the flake along the direction perpendicular to the stripes. As detailed in discussion in Supplementary Information Section III, the dynamics of the suspended flake along the $x$ direction for a fixed $\Delta T$ is ruled by an effective potential $U_{eff}(\Delta T; x)$ (Extended Data Fig. 6i–p). For $\Delta T \lesssim -1\, \text{K}$, the minimum of $U_{exp}$ is located in the middle of the gold stripe (Extended Data Fig. 6i), where the probability density distribution $P_{eff}(\Delta T; x)$ (Eq. (59)) has a sharp peak (Extended Data Fig. 6a). Upon decreasing $\Delta T$, the peak of $P_{eff}$ becomes less pronounced, though remaining localized at the centre of the gold stripe. Eventually, for $\Delta T \approx -0.1\, \text{K}$, the probability distribution develops two comparable peaks (Extended Data Fig. 6v), the second being localized at the centre of the silica stripe. For $\Delta T \approx -0.08\, \text{K}$, this second peak becomes predominant (Extended Data Fig. 6x), in agreement with previous results in literature $^{33,58,59}$.

The stiffness of the lateral Casimir–Lifshitz force trapping the flake on the gold stripe when $T \lesssim T_c$ can be quantified by measuring the effective attractive potential to which the flake is subject. This is done by calculating the probability distribution $P_{exp}(x)$ of the $x$ component of the trajectories shown in Fig. 4b,d,f,h, from which the effective potential $U_{exp}(x)$ is determined by inverting the Boltzmann factor, that is, $U_{exp}(x) = -k_B T \ln \left( P_{exp}(x) \right)$. The resulting experimental potentials (Fig. 4k, dashed lines) are then fitted to a harmonic potential $U(x) = kx^2/2$ (Fig. 4k, solid black line) with stiffness $k = 0.1\, \text{pN} \, \text{μm}^{-1}$. Comparing with the theoretical model (Extended Data Fig. 7), we see that we can obtain a value of the stiffness on the same order of magnitude for an appropriate range of the flake’s size.

Conclusions and outlook
Our method provides a novel way of controlling the distances of micro- and nanomembranes using tunable critical Casimir forces to counteract forces such as the Casimir–Lifshitz force, thereby preventing stiction and device failure. Due to the simplicity of our design—which does not require additional external actions such as that of magnetic fields—the concept can be easily adapted to already existing MEMS and NEMS. Moreover, this path opens up new possibilities for the dynamic control of MEMS and NEMS where the temperature of the system could be controlled via light illumination, enabling faster transitions and higher selectivity for a new generation of the micromembranes that are found ubiquitously in MEMS and NEMS devices.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01795-6.

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Methods
Calculation of the Casimir–Lifshitz and critical Casimir forces
The theoretical model which relates the measured experimental temperatures to the equilibrium heights of the flake in the presence of two different boundary conditions of the order parameter accounts for the following forces: the weight and buoyancy of the flake, the DLVO electrostatic interaction, the Casimir–Lifshitz force and the critical Casimir force.

Weight and buoyancy. For the hexagonal flakes used in our experiments (Fig. 2a), the sum of the weight and the buoyancy is within the range of 5–30 fN, depending on the volume of the flake. In the case of a flake suspended above an uncoated silica substrate, this sum is negligible in comparison with the Casimir–Lifshitz attraction and the electrostatic forces, the magnitude of which is within the range of 1–10 pN for distances on the order of 100–200 nm, such as those observed in our experiments. In the case of a flake suspended above an uncoated silica substrate, instead, the sum of the weight and buoyancy is not negligible compared with the other relevant forces because the flake hovers at a higher height, i.e., 5–30 fN, depending on the volume of the flake. In the case of a flake (Fig. 2a), the sum of the weight and the buoyancy is within the range of 5–30 fN, depending on the volume of the flake. In the case of a flake suspended above a gold-coated substrate, this sum is negligible in comparison with the Casimir–Lifshitz attraction and the electrostatic forces.

Electrostatic force. The DLVO electrostatic force \( F_{\text{el}} \) between the flake and the substrate, at a distance \( h \), is modelled according to Eq. (3) and is parametrized by the Debye screening length of the critical mixture \( \lambda_0 \) and the parameter with dimensions of pressure, \( P_0 \). For each choice of the surfaces involved, we fitted these parameters independently, determining the combination of values of \( \lambda_0 \) and \( P_0 \) that minimizes the \( \chi^2 \) (square error) when comparing the experimental data for the average height \( \langle h \rangle \) with the model. As a result, the various substrates turn out to be characterized by slightly different values of the parameters of the electrostatics, with a pronounced difference in the case of an uncoated surface, where the existence of larger separations between the flake and the substrate leads to larger fluctuations of \( \langle h \rangle \) (Extended Data Fig. 4). For our experiments, we found the following combinations:

| Surface Type | Parameter Values |
|--------------|------------------|
| gold-coated  | \( \lambda_D = 16 \pm 0.5 \text{ nm} \), \( P_D = 3.7 \pm 0.6 \text{ kPa} \) |
| gold-coated  | \( \lambda_D = 17 \pm 0.5 \text{ nm} \), \( P_D = 1.5 \pm 0.3 \text{ kPa} \) |
| uncoated     | \( \lambda_D = 17 \pm 0.5 \text{ nm} \), \( P_D = 28 \pm 5 \text{ kPa} \) |

As expected, these three independent fits give very similar values of \( \lambda_0 \), while the value of \( P_0 \) in the case of uncoated silica is much larger than in the case of the gold-coated substrate, indicating a stronger electrostatic repulsion.

Casimir–Lifshitz force. The Casimir–Lifshitz force \( F_{\text{CL}} \) in Eq. (1) was calculated according to ref. 49 for the layered planar bodies involved in the experimental setup in Fig. 1a. The corresponding free energy per unit area is given by:

\[
G_{\text{CL}}(h) = -\frac{A_{22}(h)}{12\pi h^2} - \frac{A_{24}(h + a)}{12m(h + a)} - \frac{A_{42}(h + b)}{12m(h + b)} - \frac{A_{44}(h + a + b)}{12m(h + a + b)},
\]

where \( A_{22}, A_{24}, A_{42} \) and \( A_{44} \) are the Hamaker functions (Supplementary Information Section I), \( a \) is the thickness of the gold layer deposited on the substrate and \( b \) is the thickness of the gold flake. In the simpler case of a bottom slide of uncoated silica, instead, the dispersion forces can be derived from

\[
G_{\text{CL}}(h) = -\frac{A_{22}(h)}{12\pi h^2} - \frac{A_{24}(h + b)}{12m(h + b)}.
\]

Critical Casimir force. The expression for the critical Casimir force \( F_{\text{crit}} \) in Eq. (2) depends on the correlation length \( \xi \) of the order parameter fluctuation of the binary liquid mixture, which is related to \( \Delta T \) via

\[
\xi = \xi_0(\Delta T/T)^{\nu_0}
\]

with \( \nu_0 \approx 0.63 \) as the critical exponent of the Ising universality class to which the mixture belongs.

Total force and potential. The total force \( F_{\text{tot}} \) acting on a flake is

\[
F_{\text{tot}}(h, \Delta T) = F_{\text{el}}(h) + F_{\text{CL}}(h) + F_{\text{crit}}(h, \Delta T) + F_{\text{w-b}}.
\]

in terms of the potentials corresponding to the forces shown in Eq. (7). In the case of a gold flake floating on a gold-coated substrate, \( U_{\text{tot}}(h, \Delta T) \) displays a very sharp minimum at a certain \( h(\Delta T) \) for all \( \Delta T \), while for the flake suspended on an uncoated silica substrate, the minimum of the potential becomes shallow for \( |\Delta T| \geq 0.1 \text{K} \).

When comparing with experiments, one has to keep in mind that the experimentally accessible quantity is the average height of the potential

\[
\langle h \rangle = \int_0^{\infty} dh \ h P(h, \Delta T),
\]

which is defined in terms of the equilibrium probability distribution function \( P(h, \Delta T) \) associated with the potential \( U_{\text{tot}}(h, \Delta T) \), that is,

\[
P(h, \Delta T) = \frac{1}{Z} e^{-\frac{U(h, \Delta T)}{\xi_0}},
\]

where \( Z = \int_0^{\infty} e^{-\frac{U(h, \Delta T)}{\xi_0}} dh \). Hence, the comparison with the experimental data must consider the equilibrium height of the potential \( h \), defined as

\[
\frac{dU_{\text{tot}}(h_{eq})}{dh} = 0.
\]
In fact, when \( \lambda_D \) becomes comparable to \( \xi \), the description of the electrostatics with Eq. (3) is no longer appropriate. Given that the estimated value of the Debye screening length is \( \lambda_D \approx 16-17 \text{ nm} \), the value of the corresponding correlation length \( \xi \) is smaller than \( \lambda_D \) for the typical temperature differences in our system up to \( \Delta T = -0.1 \text{ K} \).

We first determined the parameters of the electrostatic interaction by fitting the value of the Debye screening length \( \lambda_D \) and the prefactor \( P_0 \) such that they minimize

\[
\chi^2 = \sum_{j=1}^{N} \frac{(h_j - \langle h \rangle)^2}{\Delta h^2}.
\]

where \( \langle h \rangle \) and \( \Delta h \) were calculated for each different combination of parameters for the experimental temperature \( \Delta T \) of the experimental data \( h_j \). The fitted \( \lambda_D \) is in the range of values obtained in similar experiments.

**Experimental details**

We consider a dilute suspension of gold flakes (monocrystalline, polydisperse and with average thickness \( d = 34 \pm 10 \text{ nm} \) determined via atomic force microscopy measurements\[^5\]). The gold nanoflakes are wet-chemically synthesized using a rapid and seedless method in aqueous solution described in detail in ref. \[^5\]. Briefly, 100 \( \mu \text{L} \) of 100 \( \text{mM} \) HAuCl\(_4\) was added to 3 \( \text{mL} \) of 20 \( \text{mM} \) CTAB aqueous solution, in a glass vial, and the mixture was gently mixed and left undisturbed for several minutes. Then, 100 \( \mu \text{L} \) of 100 \( \text{mM} \) ascorbic acid was added to the mixture, followed by rapid inversions of the vial for 10 s. The resulting solution was immediately placed in a water bath at 85 \( ^\circ \text{C} \) and kept undisturbed for about 1 h. The products were washed by centrifugation at 4,000 rpm (2,218g) for 10 min and finally dispersed in deionized water for further experiments.

The flakes obtained as described above were then suspended in a near-critical binary liquid mixture of water and 2,6-lutidine at the critical composition of lutidine \( c_l = 0.286 \), having a lower critical point at \( T_c = 34 \text{ °C} \) (ref. \[^5\] and Extended Data Fig. 8). The suspension was confined in a sample cell between a microscopy slide and a cover slip spaced by about 300 \( \mu \text{m} \).

On top of the cover slip, a 40-nm-thick gold layer was sputtered homogeneously across the sample. The cover slip was then left overnight in 1 mM solution of thiols (1-octanethiol for hydrophobic and 6-mercapto-1-hexanol for hydrophilic treatment) and ethanol, creating a SAM on top of the gold. To create a hydrophobic SAM on top of the glass, trichloro(1H,1H,2H,2H-perfluorooctyl)silane was evaporated for 4 h under vacuum. The stark contrast in the resulting wetting angle \( \theta_H \), \( \theta_W \) of spurious concentrations of salt in solution, as discussed below. However, we can neglect any influence of salt in our system, as our gold flakes, originally prepared in CTAB buffer solution, have been diluted more than 2,000 times with deionized water. This is confirmed by the fact that, even after the surface treatment with SAM or in control experiments in pure water, the average height \( h = 100 \text{ nm} \) of the flake does not change (Extended Data Fig. 4).

**Hydrodynamic simulations**

In our hydrodynamics simulations, a flake is modelled as a rigid assembly of \( N \) spheres (typically, \( N = 1,000 - 3,000 \)) of radius \( r \), with \( r = b/2 \), where \( b \) is the thickness of the flake, glued together and arranged on a regular hexagon. The assembled object is immersed in an incompressible Newtonian fluid bounded by a no-slip wall. At low Reynolds numbers, the fluid dynamics is governed by the Stokes equations, in which viscous forces dominate inertial forces. Under these conditions, the translational velocity \( V \) of the sphere labelled by the index \( i = 1, ..., N \) and composing the conglomerate is linearly related to the force \( F \) exerted on either the same sphere \( (j = i) \) or on adjacent spheres via the hydrodynamic mobility tensor\^[10]. Specifically,

\[
V_i = \mu F_i + \sum_{j \neq i} \mu_{ij} F_j,
\]

where \( \mu \) denotes the self-mobility tensor of a single sphere sedimenting near a hard wall and \( \mu_{ij} \) corresponds to the pair mobility tensor quantifying the hydrodynamic interactions between the spheres and the wall\[^10\]. Here we employ the well-established Rotne–Prager approximation\[^11\] combined with the Blake solution\[^12\] to account for the corrections due to the presence of the wall. The diffusion coefficient is obtained from the drag coefficient, which, in turn, is obtained by following the usual procedure of evaluating the mean drag per sphere within the assembly. The accuracy of our approach has been assessed by close comparison of the predicted bulk drag coefficients with the exact numerical simulations of a closely packed conglomerate of spheres, which was obtained by using the freely available and open-source library HYDROLIB (Extended Data Fig. 10).

These results allow us to simulate the diffusion of a regular hexagonal flake with side length \( a \) at height \( h \) above a wall as in the representative case of a flake with \( a = 840 \text{ nm} \) in Fig. 2d (cases of particles with different sizes are shown in Extended Data Fig. 10c).

Moreover, hydrodynamic simulations allow us to investigate the influence of a possible Brownian rotation on the diffusion of the flake in the presence of the confining interface, which we find to be negligible.
As a last remark about the hydrodynamic simulation, its underlying assumption is that the viscosity of the solvent does not depend substantially on temperature. This is actually the case in experiments. In fact, the viscosity of water–lutidine at critical concentration does not change appreciably in the temperature range of our experiments. Moreover, we do not possess experimental evidence of the presence of a wetting layer.

Optical measurements and analysis
Reflection spectra at normal incidence (numerical aperture 0.5) were collected by using an inverted microscope (Nikon Eclipse TE2000-E) equipped with an oil-immersion 100× objective (switchable numerical aperture of 0.5–1.3, Nikon), directed to a fibre-coupled spectrometer (Andor Shamrock SR-303i), equipped with an Andor iDus 420 charge-coupled detector (see schematic in ref. 23). The obtained reflection spectra were analysed by the standard transfer matrix method23.

Data availability
All experimental data are available via the depository figshare under https://doi.org/10.6084/m9.figshare.20037581.v166. Source data are provided with this paper.

Code availability
The codes that support the findings of this study are available from the corresponding authors upon reasonable request.

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Acknowledgements
G.V. and F.S. acknowledge partial support from the ERC starting grant ComplexSwimmers (grant no. 677511). G.V. acknowledges support from Vetenskapsrådet (grant no. 2016-03523). A.G. acknowledges support from MIUR PRIN project 'Coarse-grained description for non-equilibrium systems and transport phenomena (CO-NEST)' no. 2017/98CZL. Fabrication in this work was partially done at Myfab Chalmers. A.D.-M.-I. gratefully acknowledges support from the Deutsche Forschungsgemeinschaft through project no. DA 2107/1-1.

Author contributions
All authors discussed the results and wrote or edited the manuscript. F.S., B.M. and G.V. planned the experiments. F.S. and B.M. performed the experiments. A.D.-M.-I. and H.L. conducted the hydrodynamic simulations. A.C. and A.G. conceptualized the theoretical model and performed the force simulations. F.S. and R.V. fabricated the experimental samples. B.M. fabricated the flakes.

Funding
Open access funding provided by University of Gothenburg.

Competing interests
The authors declare no competing interests.

Additional information
Extended data is available for this paper at https://doi.org/10.1038/s41567-022-01795-6.

Supplementary information
The online version contains supplementary material available at https://doi.org/10.1038/s41567-022-01795-6.

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Peer review information Nature Physics thanks the anonymous reviewers for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Schematics of the materials involved in the experiments. a, Configuration used to calculate the force acting on the flake depending on the material properties and on the distances indicated by the arrows. See Eqs. (1), (S1), and (S2) for the case of a gold-coated silica substrate. WL indicates the water–2,6-lutidine mixture, SiO₂ the silica, and Au the gold layers. b, Same as panel a but for the case of an uncoated silica substrate. In both panels, the text indicates each interface in the notation used for the Hamaker function (Eqs. (S1) and (S3)) and the Δₐ-b (Eq. (S2)). The order of the symbols a and b in the subscript of Δₐ-b follows the convention to determine the Casimir–Lifshitz force contribution defined in ref. 48, that is, the symbol that comes first always refers to the material that is on the outside of the interface looking toward the separating medium, which in our case is the layer of water–2,6-lutidine of thickness h. The double arrows in both panels indicate the pair of interfaces considered for each Hamaker function: in a the terms in Eqs. (S1) and b the terms in Eqs. (S3). c, Dielectric response functions of the materials involved in the experiment. For the parametrization of these functions with respect to water–2,6-lutidine and silica we follow ref. 30, while for gold we follow ref. 48, page 268, Table L2.4. Gold, Four-term fit to absorption data.
Extended Data Fig. 2 | Schematic of the experimental setup for digital video microscopy. Homemade version of an inverted brightfield microscope consisting of a white light (LED) illuminating the sample from above, which is temperature-stabilized by using a two-stage temperature controller at the sample and by indirect heating of the objective (O). The scattered light is collected by the 100 × objective (numerical aperture = 1.30) and projected onto a camera. The flake motion is tracked and analysed with digital video microscopy.
Extended Data Fig. 3 | Height measurement of a metal flake using Fabry–Pérot cavity modes. a, In a bright-field image of a gold flake, the tilt with respect to the substrate becomes visible as a change in colour, whereas in b, no colour variation is observed for a flake oriented parallel to the substrate. c, The Fabry–Pérot cavity modes are measured across the flake along the dashed lines indicated in panels a and b and reveal a shift in energy corresponding to a variation of the distance $h$ from the substrate from $h = 90 \pm 10$ to $210 \pm 10$ nm. In panel d, instead, the measured energy values remain constant and correspond to a distance $h = 90 \pm 10$ nm above the substrate. The colour bar shows the measured reflection intensity. Scale bars in panels a and b correspond to 1 μm. The analysis of the Fabry–Pérot experimental data and the determination of the local height of the cavity has been conducted according to the transfer-matrix model, which is described in ref. 23.
Extended Data Fig. 4 | Dependence of the flake height on surface treatment. Far away from $T_c$, the particle height $h$ above a gold surface is reduced to $h \approx 100$ nm due to the attractive Casimir–Lifshitz force, independent of whether a SAM is present (red circles, inset with red frame) or not (yellow diamonds, inset with yellow frame). For dielectric surfaces such as silica (grey triangles, inset grey frame), the particle fluctuates at a much larger distance $h \approx 300$ nm because of the substantially smaller attractive Casimir–Lifshitz force. Here, all measurements have been performed far away from criticality with $\Delta T \leq -0.3$ K, in a range where these results are independent of $\Delta T$. All error bars indicated in the plot correspond to the standard deviation and amount to about 5% of the absolute value. The data are presented as mean values.
Extended Data Fig. 5 | Comparison between the dispersion forces on a flake suspended on an untreated silica substrate and on a gold-coated substrate.

**a**, Casimir–Lifshitz force acting on a gold flake suspended at a height $h$ above a gold coated substrate (orange line) or an untreated silica substrate (grey line). The magnitude of the force is represented in a log-log plot and is calculated for a reference hexagonal flake with side length equal to 700 nm. The force is attractive for both substrates, but in the case of an untreated silica substrate the force is one order of magnitude smaller than in the gold-coated case. **b**, Experimental values of $\langle h \rangle$ (see Eq. (9) for its definition) of the flake for the various cases, that is, gold flake suspended on a gold-coated substrate with hydrophilic SAM (red symbols), hydrophobic SAM (blue symbols), and uncoated silica substrate (green symbols). The solid lines represent the theoretical model. The resulting fit is in good agreement with the experimental measurements for all substrates. It is worth noting that the confidence interval for the values of $\langle h \rangle$ for the uncoated silica substrate are much larger than those for the gold-coated substrates. This happens as the minimum of the total potential is very shallow for an uncoated silica substrate, because the Casimir–Lifshitz forces are much weaker in this case. At $T \ll T_c$, that is, in the absence of attractive critical Casimir forces, the gravity plus buoyancy is the only force that is effective in pushing the flake towards the substrate. In the case of a gold-coated substrate, the Casimir–Lifshitz forces are very relevant for $T \ll T_c$, and the resulting total potential has a very pronounced minimum, which is reflected in the very narrow error intervals. In the figure, the darker shade of each colour represents the 68% confidence interval. The error on the experimental data is the standard deviation from the mean of three measurements. The data are presented as mean values.
Extended Data Fig. 6 | Trapping and release of a flake, prediction of the theoretical model. a–h, Two dimensional potential $U_{tot}(\Delta T; h, x)$, i–p, effective lateral potential $U_{eff}(\Delta T; x)$, and q–x, effective lateral probability distribution function $P_{eff}(\Delta T; x)$ for a disk-like flake (radius $a = 1450 \text{ nm}$, thickness $b = 40 \text{ nm}$) hovering on a patterned substrate with hydrophobic, 3-$\mu$m-wide, gold-coated stripes alternating periodically with hydrophilic, 3-$\mu$m-wide, uncoated silica stripes. The thickness of each gold-coated stripe is $a_1 = 30\text{ nm}$. As shown in panels a–h, the height of the flake $h$ on the surface is measured with respect to the upper surface of the gold stripes; hence, when considering the height of the flake with respect to the hydrophilic uncoated surface, one must take into account the additional term $a_1$. The profile of the substrate is represented in panels a–h. The value of the two-dimensional total potential is represented with a contour plot. For $\Delta T = -1$ K, the minimum of the total potential is located over the gold-coated stripe at its centre (for symmetry reasons) about 100nm above the surface. Upon increasing the temperature towards $T_c$, the minimum of the potential becomes less and less deep, and for $\Delta T > -0.1$ K, the minimum is located above the uncoated silica stripe. In i–p, the effective lateral potential $U_{eff}(\Delta T; x)$ defined in Eq. (S8) is represented with a black continuous line. The zero of the potential is set at $x = 0$. For $\Delta T \leq -0.12$ K (i–m), the effective potential has a single minimum at $x = 1.5 \mu$m; for $\Delta T > -0.1$ K (o–p), the effective potential has a single minimum at $x = 4.5 \mu$m, that is, localized at the centre of the uncoated silica stripe; for $\Delta T = -0.1$ K (n), the effective potential has two local minima. In i–p, the effective probability distribution $P_{eff}(\Delta T; x)$ defined in Eq. (S9) is represented with a black continuous line. In i–m, $P_{eff}(\Delta T; x)$ has a single peak at $x = 1.5 \mu$m, indicating that the flake is localized on the gold stripe. The peak is initially very sharp, but becomes broader and less high upon increasing the temperature towards $T_c$. In n, $P_{eff}(\Delta T; x)$ has two peaks. In n, o, the peak is again one, this time localized at the centre of the uncoated silica stripe. In this model, the parameters for the electrostatic interaction are $\lambda_D = 17 \text{ nm}$ and $P_0 = 1.5 \text{ kPa}$ for the gold-coated stripes, whereas $P_0 = 28.8 \text{ kPa}$ for the gold-coated stripes.
Extended Data Fig. 7 | Effective stiffness predicted by the theoretical model. Stiffness of the lateral trapping ($k_x$) calculated for the effective potential $U_{\text{eff}}$ as a function of $\Delta T = T - T_c$. Here we calculate $k_x$ using the variance of the peak of the probability distribution $P_{\text{eff}}(\Delta T; x)$, that is, $k_x = \frac{k_B T}{\sigma^2}$ where

$$\sigma^2 = \int_{x_1}^{x_2} [P_{\text{eff}}(\Delta T; x) (x - x_{\text{peak}})]^2 \, dx.$$ 

The extrema of the integral are defined as $x_1 = 0, x_2 = 3 \, \mu\text{m}$ for the $\Delta T$s where the absolute peak of $P_{\text{eff}}(\Delta T; x)$ falls in the centre of the gold-coated stripe, and as $x_1 = 3 \, \mu\text{m}, x_2 = 6 \, \mu\text{m}$ for the $\Delta T$s where the absolute peak of $P_{\text{eff}}(\Delta T; x)$ falls in the centre of the uncoated silica stripe. We remark that, for our model, $\sigma$ is determined not only by the parameters of the interaction, but also by the geometrical characteristics of the system. In particular, the main factor affecting $\sigma$ in our model is the difference between the diameter of the flake and the width of the stripes. In fact, $U_{\text{eff}}(\Delta T; x)$ is necessarily flat over an interval of length $w - 2R$, where $w$ is the thickness of the stripe and $R$ is the radius of the disk-like flake. Hence, for the same parameters describing the interaction per unit area, different flake dimensions have a $\sigma$ that is at least $w/2 - R$, which increases upon decreasing $R$. 

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Extended Data Fig. 8 | Phase diagram of a water–2,6-lutidine liquid mixture. The binary solution of water and 2,6-lutidine possesses two distinct phases given by the binodal (black line) and spinodal line (black dotted line) separating mixed from demixed phases. The solution is prepared at $T_0 = 32.5\,^\circ\text{C}$ and the critical concentration of 2,6-lutidine $c_c = 0.286$ near the critical point (CP) with $T_c \approx 34\,^\circ\text{C}$. The data are obtained from ref. 61.
Extended Data Fig. 9 | Effect of the treatment with different self-assembled monolayers (SAM). Side and top view of a cover glass slide half of which has been sputtered with a 30 nm gold layer (left half). The treatment with hydrophilic thiols attached to gold and hydrophobic silanation to glass is visible by the difference of the wetting angle (black dashed line in the side view) and by the spreading (black dashed circle in the top view) of a 10 μL droplet.
Extended Data Fig. 10 | Scaled diffusion coefficient for edgewise translational motion of a flake composed of a conglomerate of spheres arranged on a regularly shaped hexagon in an unbounded (bulk) fluid or close to a planar hard wall. **a**, Comparison between the Rotne–Prager approximation employed in the present work (red squares) and the bulk diffusion coefficients obtained using the open-source software HYDROLIB (blue circles). **b**, The predictions for the scaled diffusion coefficient are obtained by using the Rotne–Prager approximation combined with the Blake solution for hydrodynamic interactions between the spheres near a hard wall. Three flake sizes of aspect ratio 16, 20, and 35 are considered, which correspond to total numbers of spheres of 817, 1261, and 3781, respectively. Horizontal dashed lines correspond to the scaled diffusion coefficient in an unbounded fluid medium, that is, in the absence of the confining wall given by the substrate. **c**, Hydrodynamic simulations of a regular hexagon with various particle sizes: simulated diffusion constant $D$ for hexagonal particles with side length $a = 700$, $840$, $1000$, $1450$ nm depending on their distance to the surface $h$. Solid lines represent the fitting of the simulation data. See Supplementary Information Section IV for additional details.