Van der Waals forces are ubiquitous across the sciences, being critical to adhesion, friction and the stability of colloids. The interaction has the same origin as Casimir forces, meaning it can be understood in terms of the exchange of virtual photons between atoms or molecules. Such an interaction usually pulls the interacting particles together, and in some special circumstances can push them apart. However, no method for engineering the van der Waals force to be perpendicular to the inter-particle separation vector has been predicted or observed. Here we show that such a force can exist in realistic laboratory conditions, by inducing a circular dipole moment in one of the atoms. The mechanism is similar to the spin-orbit coupling that leads to lateral atom-surface forces, but here the angular momentum in a single excited atom leads to a torque acting on the two-atom system. The forces predicted here provide a new tool for optomechanical manipulation of bound systems which will have far-reaching relevance across micro and nano electromechanical systems (MEMS/NEMS), as well as in colloidal and atomic physics.

Casimir forces [1–3] are fluctuation-induced interactions between polarisable macroscopic objects, which can be ascribed to zero-point fluctuations of the electromagnetic field. They act over very short distances, but scale with large inverse powers of the separation, so that they quickly go from negligible to dominant as distances are continually reduced in nanomechanical devices. Lateral Casimir forces (those perpendicular to the separation vector of the objects involved) have recently become a focus of research due to their potential to realise contactless force transmission, as well as novel types of sensors and clocks [4–7]. Traditionally, lateral Casimir (surface–surface) and Casimir–Polder (atom–surface) forces have been predicted [8–11] and measured [12, 13] by considering corrugated surfaces [14–19].

More recently it has been recognised that a lateral force can also be achieved by placing a circular emitter near a featureless planar surface [20–27] or nanofiber [28–34] as depicted in Figs 1a and 1b. The required breaking of mirror symmetry is provided by the circular emitter’s angular momentum vector, which is aligned along a particular direction. Spatial inversion along this vector then causes the emitter’s rotation to reverse. This scheme is easily experimentally realisable — for example a Zeeman sublevel of the atom can be irradiated with light carrying orbital angular momentum, resulting in a circular dipole. The atom then excites strongly-directional guided modes in a nearby surface, which couple to its angular momentum — a type of spin-orbit coupling where the light polarisation is correlated with its propagation direction in a non-trivial manner [35–38]. Conservation of momentum dictates that the emitter will be propelled in a direction opposite that of stronger emission. Lateral forces of this kind are unbounded in the sense that they can be used to continuously propel an atom along a surface. By contrast, lateral forces due to gratings are conservative and only act until the atom has reached a potential minimum above a grating bar.

We want to transfer the idea of lateral dispersion forces to the context of molecular and colloidal physics by identifying the simplest and most elementary system that permits such an effect. This will at the same time illuminate the microscopic origin of lateral atom–surface forces. To this end, we investigate the van der Waals force between two isolated atoms, where one is excited and has a circular dipole moment while the other’s dipole moment is isotropic. As indicated in Figs. 1c
and 1d, we will show that there is a force perpendicular to the inter-atomic separation acting on the excited, circularly polarised atom, which can be interpreted via photon recoil associated with anisotropic emission. It is crucial to note that this lateral force only arises when the second atom is present, since it opens a channel into which asymmetric emission can occur. In the remainder of this article we will develop the theory of such a two-atom system, quantify the orders of magnitude of the lateral force, and outline the main features of a possible experimental detection scheme.

We begin from previous results concerning for the Casimir-Polder forces between and atom and a surface. As shown in [40] and in the Supplementary Material, the resonant Casimir-Polder force $F_A$ on an atom at position $r_A$ at time $t$ reads:

$$F(r_A, t) = 2\mu_0 p_1(t) \omega_A^2 \text{Re} \left\{ \nabla [d_{10}^A \cdot G(r, r_A, \omega_A) \cdot d_{01}^A] \right\}_{r=r_A},$$  \hspace{1cm} (1)

where $\mu_0$ is the permeability of free space, $\omega_A$ is the transition frequency, $d_{10}^A$ is the transition dipole moment between atomic states 0 and 1, and $G(r, r', \omega)$ is the classical Green’s tensor (or propagator), which describes the amplitude for the propagation of a photon between two points in space and time, subject to boundary conditions set by the medium. We have assumed that the atom A is prepared in an energy eigenstate $|1\rangle$ with population $p_1(t)$. We can interpret formula (1) in simple terms by reading from right to left. The atom initially emits a photon, changing its state from $|1\rangle$ to the intermediate state $|0\rangle$. The photon is then reflected by the surface (a process described by the Green’s tensor $G$) and finally is reabsorbed by the atom via the dipole moment $d_{10}^A$.

Our next step is to recognise that the Casimir–Polder force can also be understood as a recoil force associated with an irreversible rate of excitation transfer from the initially excited atom to its surroundings. To investigate this further in the context of lateral forces, we consider a plate which breaks the translational symmetry in one direction (chosen as the $z$ axis), but preserves it in $x$ and $y$, allowing us to Fourier transform the Green’s tensor along those directions: $G(r, r', \omega) = \int d^2k || e^{ik || (r-r')} G(z, z', \omega, k ||) $, where $k || \cdot \hat{z} = 0$. Furthermore, instead of directly considering the force as given by Eq. (1), we will uncover its microscopic origin by considering the emission rate $\Gamma$ of an atom prepared in eigenstate $|1\rangle$ near such a medium. As shown in the Supplementary Material, this is given by

$$\Gamma(z_A) = \int d^2k || \frac{2\mu_0}{\hbar} \omega_A^2 \text{Im} \left\{ d_{10}^A \cdot G(z_A, z_A, \omega_A, k ||) \cdot d_{01}^A \right\}$$

$$= \int d^2k || \gamma(z_A, k ||),$$  \hspace{1cm} (2)

where we have defined a rate density $\gamma$ for emission of photons of lateral momentum $h k ||$. Direct comparison of Eqs (1) and (2) reveals that

$$F_z(z_A, t) = - \int d^2k || h k || p_1(t) \gamma(z_A, k ||),$$  \hspace{1cm} (3)

where $x$ is a direction parallel to the surface, so that $F_z(z_A, t) = F_A(z_A, t) \cdot \hat{x}$. This formula can then be directly interpreted as a recoil force originating from an emission rate density $\gamma$ describing directional radiation by the atom, with the minus sign accounting for the fact that the recoil force is opposite to the direction of emission. Clearly, if the rate density $\gamma$ used in the above equation is asymmetric with respect to the $x$ axis, the integral is non-zero, giving a force along $x$.

So far we have taken the known atom-surface force (1), and reinterpreted it in terms of a rate (2). We now replace the surface with a single isotropically polarised atom, so that the forces involved are no longer Casimir–Polder (atom–surface), but van der Waals (atom–atom). We make this replacement by using the dilute-gas limit considered by Lifshitz [3]. The main idea is that, from a microscopic point of view, a macroscopic medium is a collection of a large number of atoms — the imposition of macroscopic boundary conditions is simply a neat and powerful way of summarising their collective behaviour. This means that the van der Waals interaction should be contained within the Casimir–Polder interaction (1). This is indeed the case, since the Green’s tensor admits a Born expansion for a dilute body [47]:

$$G(r, r', \omega) = G(0)(r, r', \omega)$$

$$+ \mu_0 \omega^2 \int d^3r_B \rho(r_B) G(0)(r, r_B, \omega) \cdot \alpha_B(\omega) G(0)(r_B, r', \omega)$$  \hspace{1cm} (4)

where $\rho$ is the number density of the medium, $G(0)(r, r', \omega) = 1 + (\epsilon^2/\omega^2) \nabla \nabla |e^{i\omega|r-r'|/c}|^2/(4\pi |r-r'|)$ is the free-space Green’s tensor and $\alpha_B(\omega)$ is the isotropic polarizability of the medium’s constituent atoms, each prepared in the ground-state. In Eq. (4) first term $G(0)(r, r', \omega)$ represents free propagation of the photon from the initial position $r'$ to $r$, while the integral term represents an environment-modified propagation. We can interpret the terms in the integrand of (4) by reading from right-to-left in a similar fashion to Eq. (1). In the rightmost term the photon propagates freely from $r'$ to an atom of the medium placed at $r_B$, then polarises the atom according to $\alpha_B$. Then, in the leftmost term the photon is scattered to its final position $r$.

Using this Born expansion we see that the Casimir-Polder force between atom A and the dilute medium can be understood as the pairwise summation of van der Waals forces between atom A and the constituent atoms of B: $F_A(r_A, t) = \int d^3r_B \rho(r_B) F(r_A, r_B, t)$. In particular the resonant contribution to the atom-atom interaction
\[ F_A(r, t) = \text{Re} \left\{ \nabla \left[ d_A^A \cdot G^{(0)}(r, r, \omega_A) \cdot \alpha^B(\omega_A) \right] \right\} = \frac{\pi p_1(t) \omega_A^4}{8 \pi \varepsilon_0 \varepsilon_r} \sin \left( \frac{\pi}{2} \left( 3 - \xi^2 \right) \cos(2\xi) - (9 - 15\xi^2 + \xi^4) \sin(2\xi) \right) \] 

where \( \xi = \omega_A t/c, \) and \( p_1(t) \) represents the population of the excited state, whose evolution is governed by the spontaneous decay rate \( \Gamma, \) so that in the lowest order of approximation \( p_1 = e^{-\Gamma t}. \) This means that the lateral force has a population-induced dynamics reaching a steady state at large times, corresponding to the emitter having decayed to the ground-state. Naturally the force is stronger for short times. Even though we only have a single emitter, this result can be understood from the ensemble-average origin of the force: the probability of photon emission (and hence the size of the recoil) is highest for small times, which is when a large fraction of the ensemble atoms are still in their excited state.

In order to explain the microscopic origin of the lateral force (8), we calculate the emission rate density \( \gamma \) as defined in Eq. (2) using the Born-expanded Green’s tensor (4). Since \( \gamma \) describes the rate of emission of a single photon of a particular \( k ||, \) we define the following ‘recoil rate’

\[ R(r, \varphi) = \int_0^\infty d\xi \sin^2(\varphi) \gamma \left( r_A, r_B, k || \right) \] 

which describes the total rate of emission along a particular direction \( \varphi \) as defined in Fig. 2a, weighted with the associated recoil. We find the following recoil rate in the near-field (\( \xi \ll 1 \)) limit:

\[ R(r, \varphi) = \frac{d_A^A}{64 \pi^3 \varepsilon_0^2 \varepsilon_r} \alpha^B(\omega_A) \left\{ 16 \xi^3 \sin^2(\varphi) + \frac{\pi \xi^4}{8} \left[ 7 + 3 \cos(2\varphi) \right] + \frac{2\xi^5}{15} \left[ 35 + 24 \cos(\varphi) - 3 \cos(2\varphi) \right] \right\}. \]

Because of the term proportional to \( \cos \varphi, \) the excitation of radiation modes that propagate into the \( +x \) or \( -x \) direction is in general asymmetric, as shown in Fig. 2a where we plot the asymmetry of the emission spectrum for the excited caesium atom. Unidirectional mode excitation is observed for the scattering of light by the circularly polarised emitter, with a stronger emission in the negative \( x \) direction resulting in a force in the positive \( x \) direction, as shown in Fig. 2b.

If the atom begins from rest, after a time of the order \( \Gamma^{-1} \) its velocity due to this force has an order of magnitude of \( F_{\lambda, x}(r, 0)/(m\Gamma) \) since \( p_1(t) = e^{-\Gamma t}. \) This turns out to be rather small, so a more convenient experimental setup is one where a continuous laser driving with Rabi frequency \( \Omega \) is used. In the case of large laser detuning \( \Delta \) and weak excitation (\( \Gamma \ll \Omega \ll |\Delta| \), the steady state population of the excited state is given by \( p_1(t \gg 1/\Gamma) = \Omega^2/4\Delta^2. \) For example if \( \Omega = 0.2 |\Delta| \) the excited population is \( p_1(t \gg 1/\Gamma) = 10^{-2} \) and the velocity \( v \) gained due to application of this force has an order of magnitude of \( F_{\lambda, x}(r, 0)p_1(t \gg 1/\Gamma) \Delta t/m, \)
FIG. 2. Panel (a) shows the emission spectrum of an excited caesium atom at a distance \( r = 632 \text{nm} \) an isotropically polarised rubidium atom. This distance corresponds to the first peak in the plot (b), which shows the lateral force on the caesium atom. In panel (b) we also show the emission spectrum a selection of separations \( r \), it is seen that the emission is symmetrical for the points at which there is zero lateral force, as expected. The vertical axis on the right hand side is the velocity which the atom gains if continuously excited with a laser driving, as explained in the text. The inset shows the small-distance behaviour of the force, with all axes having the same units as in the main plot. Finally in panel (c) we sketch the effect of the lateral force on a molecular system, which would be to induce a torque on the whole sample.

where \( \Delta t \) is the time over which the laser is applied. Figure 2b shows the lateral force for short times and the mean velocity gained during the thermalisation process, where we have taken \( \Delta t = 10 \text{ms} \). At distances of, say, \( r = 0.1 \mu \text{m} \), wave function overlap between the two species can safely be neglected and the lateral velocity acquired by the caesium atom is around 800nm/s as shown in the inset of Fig. 2b.

Both the force and the velocity show characteristic Drexhage-type oscillations [41] if the distance \( r \) between the two atoms is varied. This means that we can selectively excite the radiation modes in the positive or negative \( x \) direction at will. At very small distances the force diverges, but this is to be expected as the formalism used here only applies when there is no appreciable wave function overlap between the two atoms, which happens at distances of the order a few angstroms.

The lateral force acting on the ground-state atom is vanishing as shown in the Supplementary Material, meaning that the lateral force induces a torque on a two-atom sample, as sketched in Fig. 2c. There the ever-present longitudinal forces are also indicated. As shown by [49–51], the longitudinal forces \( F_{A,z} \) acting on each atom are not equal and opposite due to the momentum carried by the photon. Hence the center-of-mass of the whole system is subjected to a translational acceleration, superimposed with the twisting motion induced by the lateral force derived here. When both atoms have reached thermal equilibrium the recoil forces vanish and the center-of mass has an uniform motion, as shown in the upper part of Fig. 2c. Selectively exciting one atom in a dimer could allow one to observe the translational and rotational degrees of freedom in the molecular motion.

To conclude, we have demonstrated the existence of a lateral van der Waals force between an excited, circularly polarised atom and an isotropic atom. This could be experimentally realised by selectively pumping the atom to a Zeeman sublevel with a maximal \( y \)-component of angular momentum. Control of the lateral force direction and magnitude can be experimentally implemented by changing the handedness of the illuminating light and the distance between the two atoms. Our work is the
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I. SUPPLEMENTARY MATERIAL

In the main article we concentrated on the van der Waals force acting on the excited atom. It is the aim of the supplementary material to consider also the force acting on the ground-state atom. In order to derive this quantity we can consider the Casimir–Polder force acting on a ground-state atom near an excited medium, a setup previously investigated in the literature \[61, 62, 64\]. To be more general, we will consider the case that both the atom and the medium are excited and generalise the Born expansion to this case.

A. From rate to resonant forces

To compute the potential we perturbatively calculate the S-matrix elements and define the T-matrix by the equation \[63\]:

\[
S_{fi} = \delta_{fi} - 2\pi \delta(E_f - E_i) T_{fi} / \pi
\]

The real and imaginary parts of the T-matrix can be identified as the energy shift and the inverse lifetime (decay width) of the particle: \(T_{ii} = U(r_A) - i\hbar\Gamma / 2\). Hence the interaction energy reads:

\[
U(r_A) = -\frac{i}{4\pi} \int_0^\infty d\omega Tr \{ \hat{T}[\hat{H}_{AF}(t) \hat{H}_{AF}(t')] \} + c.c.
\]

where \(\hat{H}_{AF} = -\hat{d}^A \cdot \hat{E}(r_A)\) is the interaction Hamiltonian in the electric-dipole approximation, \(T\) is the time-ordered product and \(\hat{\rho}\) the atom/field density matrix. If the atom is in a incoherent superposition of energy eigenstates \(|n_A\rangle\) with occupation \(p_n^A\) and the field is in the ground-state, the density matrix reads \(\hat{\rho} = \hat{\rho}_A \otimes \hat{\rho}_F\) where:

\[
\hat{\rho}_A = \sum_n p_n^A |n_A\rangle \langle n_A|, \quad \hat{\rho}_F = |\{0\}\rangle \langle \{0\}|
\]

The time-ordering operator \(T\) moves later time arguments to the left. Using the definition of the field propagator we easily find that:

\[
U(r_A) = \frac{i\hbar \mu_0}{4\pi} \int_0^\infty d\omega Tr \{ \mathbf{D}_F(r_A, r_A, \omega) \cdot \hat{\alpha}^A(\omega) \} + c.c.
\]
where $\hat{\alpha}^A (\omega)$ is an effective polarizability in the incoherent state:

$$\hat{\alpha}^A (\omega) = \sum_n p_n^A (t) \hat{\alpha}_n^A (\omega) = \frac{1}{\hbar} \lim_{t \to 0^+} \sum_n p_n^A (t) \sum_k d_{kn}^A d_{nk}^A \left( \frac{1}{\omega_{kn}^A - \omega - i\varepsilon} + \frac{1}{\omega_{kn}^A + \omega + i\varepsilon} \right)$$

(15)

and $D_F$ is the field propagator:

$$D_F (x, x') = \frac{i}{\hbar \mu_0} \langle \{0\} | T \{ \hat{E} (r, t) \hat{E} (r', t') \} | \{0\} \rangle .$$

(16)

In general:

$$D_F (r, r', -\omega) = D_F (r, r', \omega),$$

$$D_F (r, r', \omega) = D_F^T (r', r, \omega)$$

(17)

If the atom is in the state $|n^A\rangle$ the rate which is proportional to the imaginary part of the T matrix reads [61, 62, 64]:

$$\Gamma_n^A (r_A) = -\frac{\mu_0}{2\pi} \int_0^\infty d\omega \text{Tr} \{ D_F (r_A, r_A, \omega) \cdot \hat{\alpha}_n^A (\omega) \} + \text{c.c.}$$

(18)

where $\hat{\alpha}_n^A (\omega)$ is an effective polarizability in the state $|n^A\rangle$:

$$\hat{\alpha}_n^A (\omega) = \frac{1}{\hbar} \lim_{t \to 0^+} \sum_k d_{kn}^A d_{nk}^A \left( \frac{1}{\omega_{kn}^A - \omega - i\varepsilon} + \frac{1}{\omega_{kn}^A + \omega + i\varepsilon} \right).$$

(19)

Note that the effective polarizability is different from the usual retarded polarizability in the atomic state $|n^A\rangle$:

$$\alpha_n^A (\omega) = \frac{1}{\hbar} \lim_{t \to 0^+} \sum_k \left( \frac{d_{kn}^A d_{kn}^A}{\omega_{kn}^A - \omega - i\varepsilon} + \frac{d_{nk}^A d_{nk}^A}{\omega_{kn}^A + \omega + i\varepsilon} \right)$$

(20)

As shown in the main text resonant forces can be understood as recoil forces associated with an irreversible rate of excitation transfer from the excited atom to the electromagnetic bath. Using the same arguments as above Eq. (3) in the main text, we find the following Casimir–Polder force:

$$\mathbf{F} (r_A) = -\frac{i\hbar \mu_0}{2\pi} \int_0^\infty d\omega \nabla \text{Tr} \{ D_F (r, r_A, \omega) \cdot \hat{\alpha}^A (\omega) \} |_{r=r_A} + \text{c.c.}$$

(21)

It is tempting to derive the force by just taking the gradient of the energy shift, which is the real part of the T-matrix. However our work recognises that in general Casimir–Polder forces are not conservative and cannot derived from a potential as suggested also in Refs [30, 40].

B. Atom-surface interaction

We consider first the Casimir–Polder interaction of the atom a near a macroscopic medium by calculating the propagator of the electromagnetic field using macroscopic quantum electrodynamics. We introduce the vector field $\hat{f}_\sigma (r, \omega)$ as the bosonic annihilation operator for electric ($\sigma = e$) and magnetic ($\sigma = m$) excitations. Denoting by $|\{0\}\rangle$ the vacuum state of the electromagnetic field, $\hat{f}_\sigma (r, \omega) |\{0\}\rangle = 0$, a one-photon excited state is given as $\hat{f}_\sigma (r, \omega) |\{0\}\rangle = |1_\sigma (r, \omega)\rangle$. For zero temperature the only non-vanishing thermal average of two bosonic operators is $\langle \{0\} | \hat{f}_\sigma (r, \omega) \hat{f}_\sigma^\dagger (r', \omega') | \{0\} \rangle = \delta_{\sigma \sigma'} \delta (r - r') \delta (\omega - \omega')$. Introducing the frequency components of the fields via $\hat{E} (r) = \int_0^\infty d\omega \hat{E} (r, \omega) + \text{H.c.}$, we may write the electric field in terms of the bosonic annihilation operators as

$$\hat{E} (r, \omega) = \int d^3 r' \sum_{\sigma = e, m} \mathbf{G}_\sigma (r, r', \omega) \cdot \hat{f}_\sigma (r', \omega)$$

(22)
with the tensors $\mathbf{G}_\sigma$ being defined in terms of the Green’s tensor $\mathbf{G}$ according to Ref. [46, 60]. The Green’s tensor contains all geometrical as well as magneto-dielectric properties of the environment via the relative susceptibility. It satisfies a useful integral relation with the mode-tensors $\mathbf{G}_\sigma$:

$$
\sum_{\sigma = e,m} \int d^3s \mathbf{G}_\sigma (r, s, \omega) \cdot \mathbf{G}_\sigma^{*\top} (r', s, \omega) = \frac{\hbar \mu_0}{\pi} \omega^2 \text{Im} \mathbf{G} (r, r', \omega).
$$

(23)

where $\mu_0$ is the vacuum permeability. Using the expansion of the electric field (22), the integral relation (23), we find that for zero-temperature the propagator (16) is proportional to the classical Green’s tensor ($\omega > 0$):

$$
\mathbf{D}_F (r, r', \omega) = \int_{-\infty}^{\infty} d\tau e^{i\omega \tau} \mathbf{D}_F (x, x') = \omega^2 \mathbf{G} (r, r', \omega)
$$

(24)

with the $\omega < 0$ version being found from Eq. (17). Rotation of the integration contour to the imaginary axis leads to the following non-resonant and resonant terms for non identical systems:

$$
\mathbf{F}^\text{n\tau}_A (r_A, t) = -\frac{\hbar \mu_0}{\pi} \int_0^\infty d\xi \xi^2 \text{Re} \left\{ \nabla \text{Tr} \left\{ \mathbf{G} (r, r_A, i\xi) \cdot \mathbf{\tilde{\alpha}}^A (i\xi) \right\} \right\}_{r = r_A}
$$

(25)

$$
\mathbf{F}^r_A (r_A, t) = 2\mu_0 \sum_n p^A_n (t) \sum_{k < n} (\omega^A_{nk})^2 \text{Re} \left\{ \nabla \mathbf{d}^A_{nk} \cdot \mathbf{G} (r, r_A, \omega^A_{nk}) \cdot \mathbf{d}^A_{kn} \right\}_{r = r_A},
$$

(26)

which are in agreement with recent results [30, 40].

C. Van der Waals interaction

For a dilute body the propagator admits the Born expansion [61, 62]:

$$
\mathbf{D}_F (r, r', \omega) = \omega^2 \mathbf{G}^{(0)} (r, r', \omega) + \mu_0 \omega^4 \int d^3r_B \rho (r_B) \left\{ \mathbf{G}^{(0)} (r, r_B, \omega) \cdot \mathbf{\Pi}^B_1 (\omega) \cdot \mathbf{G}^{(0)} (r_B, r', \omega) \right. \\
+ \mathbf{G}^{(0)} (r, r_B, \omega) \cdot \mathbf{\Pi}^B_{12} (\omega) \cdot \mathbf{G}^{(0)*} (r_B, r', \omega) \right\}
$$

(27)

where $\rho$ is the number density of the atoms of medium B, $\mathbf{G}^{(0)}$ is the free-space Green tensor and * indicates the complex conjugate. Moreover we have defined the polarization operators:

$$
\mathbf{\Pi}^B_1 (\omega) = \frac{1}{\hbar} \lim_{\varepsilon \to 0^+} \sum_l p^B_l (t) \sum_p \mathbf{d}^B_{pl} \mathbf{d}^B_{lp} \left( \frac{1}{\omega^B_{pl} - \omega - i\varepsilon} + \frac{1}{\omega^B_{pl} + \omega + i\varepsilon} \right),
$$

$$
\mathbf{\Pi}^B_{12} (\omega) = \frac{2\pi i}{\hbar} \sum_l p^B_l (t) \sum_p \mathbf{d}^B_{pl} \mathbf{d}^B_{lp} \delta (\omega - \omega^B_{lp}).
$$

(28)

This formula generalizes the Born expansion for a passive dilute body to the case where the atoms of the body are excited. Using this Born expansion we see that the Casimir-Polder force between the atom and the surface could be understood as the pairwise summation of van der Waals forces between the atom and the atoms of the body, $\mathbf{F} (r_A, t) = \int d^3r_B \rho (r_B) \mathbf{F} (r_A, r_B, t)$, as mentioned in the main text. Using these in Eq. (21) and rotating the integral to the imaginary axis leads to the following non-resonant and resonant terms for non identical systems:

$$
\mathbf{F}^\text{n\tau}_A (r_A, r_B, t) = \frac{\hbar \mu_0^2}{\pi} \int_0^\infty d\xi \xi^4 \text{Re} \left\{ \nabla \text{Tr} \left\{ \mathbf{\tilde{\alpha}}^A (i\xi) \cdot \mathbf{G}^{(0)} (r, r_B, i\xi) \cdot \mathbf{\tilde{\alpha}}^B (i\xi) \cdot \mathbf{G}^{(0)} (r_B, r_A, i\xi) \right\} \right\}_{r = r_A},
$$

(29)

$$
\mathbf{F}^r_A (r_A, r_B, t) = 2\mu_0^2 \sum_n p^A_n (t) \sum_{k < n} (\omega^A_{nk})^4 \text{Re} \left\{ \nabla \mathbf{d}^A_{nk} \cdot \mathbf{G}^{(0)} (r, r_B, \omega^A_{nk}) \cdot \mathbf{\tilde{\alpha}}^B (\omega^A_{nk}) \cdot \mathbf{G}^{(0)} (r_B, r_A, \omega^A_{nk}) \cdot \mathbf{d}^A_{kn} \right\}_{r = r_A}
$$

$$
+ 2\mu_0^2 \sum_l p^B_l (t) \sum_{p < l} (\omega^B_{lp})^4 \text{Re} \left\{ \nabla \mathbf{d}^B_{lp} \cdot \mathbf{G}^{(0)*} (r_B, r_A, \omega^B_{lp}) \cdot \mathbf{\tilde{\alpha}}^A (\omega^B_{lp}) \cdot \mathbf{G}^{(0)} (r, r_B, \omega^B_{lp}) \cdot \mathbf{d}^B_{pl} \right\}_{r = r_A}
$$

(30)
Because of the resonant contribution the Newtonian action–reaction balance is violated when looking at the two-atom system in isolation: \( F_A \neq -F_B \). For example if \( A \) is in the excited state and \( B \) is in the ground state \( p_B^B(t) = \delta_{B0} \) the resonant forces acting on both atoms are:

\[
F_A^A(r_A, r_B, t) = 2 \mu_0^2 \sum_n \rho_n^B(t) \sum_{k<n} (\omega_{nk}^A)^4 \text{Re} \left\{ \nabla d_{nk}^A \cdot G^{(0)}(r, r_B, \omega_{nk}^A) \cdot \alpha^B(\omega_{nk}^A) \cdot G^{(0)}(r_B, r_A, \omega_{nk}^A) \cdot d_{kn}^A \right\} \Big|_{r=r_A}, \tag{31}
\]

\[
F_B^B(r_A, r_B, t) = 2 \mu_0^2 \sum_n \rho_n^A(t) \sum_{k<n} (\omega_{nk}^B)^4 \text{Re} \left\{ \nabla d_{nk}^B \cdot G^{(0)*}(r_A, r_B, \omega_{nk}^B) \cdot \alpha^B(\omega_{nk}^B) \cdot G^{(0)}(r, r_A, \omega_{nk}^A) \cdot d_{kn}^B \right\} \Big|_{r=r_B}, \tag{32}
\]

as discussed in the main text. In particular when looking at the caesium-rubidium system, analyzed in the main text, we can see that the lateral resonant force vanishes for the ground-state atom and is not vanishing for the excited atom. The interpretation of both resonant forces is shown in Fig. 3, where frequency arguments have been dropped for brevity.

![Diagram](image)

**FIG. 3.** Resonant van der Waals forces. In the diagram on the left [corresponding to Eq. (31)] a real photon emitted by \( A \) travels to \( B \) (described via \( G(r_A, r_B) \)), excites it, then travels back to \( A \). Hence Eq. (31) is related to a reversible exchange of excitation (pendulation). The interpretation of the diagram on the right [corresponding to Eq. (32)] is more subtle. The term \( G^*(r_A, r_B) \) retains its interpretation from the left diagram, however the term \( G^*(r_A, r_B, \omega_{nk}) = G(r_A, r_B, -\omega_{nk}) \) describes propagation of a real photon of positive energy from \( B \) to \( A \) but backward in time. Hence the form shown in Eq. (32) is related to an effectively irreversible (Forster) excitation transfer due to atom \( B \)'s exposure to the field of atom \( A \); in other words the electric field of atom \( A \) causes a quadratic Stark shift on atom \( B \). This interpretation is borne out in evaluation of Eqs. (31) and (32) – the former exhibits oscillatory position dependence, while the latter is monotonic.

### D. Emission spectrum

In this section we investigate the connection between resonant forces and photon recoil, for which we need the Green’s tensor in free space, which reads:

\[
G^{(0)}(r, r', \omega) = \left( 1 + \frac{c^2}{\omega^2} \nabla \nabla \right) e^{i\omega |r-r'|/c} \frac{1}{4\pi |r-r'|} \tag{33}
\]

In order to investigate the angular dependence of the Green’s tensor, we make use of the following expansion in cylindrical coordinates:

\[
e^{i\omega |r-r'|/c} \frac{1}{4\pi |r-r'|} = \frac{i}{8\pi^2} \int_0^{2\pi} d\phi \int_0^\infty dk_\perp \frac{1}{k_\perp} e^{ik_\perp (\Delta x \cos \phi + \Delta y \sin \phi)} e^{ik_\perp |\Delta z|} \tag{34}
\]

where \( \Delta x = x - x' \), \( \Delta y = y - y' \), \( \Delta z = z - z' \) and \( k_\perp \) is the transverse momentum in vacuum:

\[
k_\perp = \sqrt{\omega^2 - k_\parallel^2}. \tag{35}
\]

This means that the Green’s tensor can also be expanded in cylindrical coordinates:

\[
G^{(0)}(r, r', \omega, k_\parallel) = \int_0^{2\pi} d\phi \int_0^\infty dk_\parallel k_\parallel G^{(0)}(r, r', \omega, k_\parallel) \tag{36}
\]

where:

\[
G^{(0)}(r, r', \omega, k_\parallel) = \frac{i}{8\pi^2} \left( 1 + \frac{c^2}{\omega^2} \nabla \nabla \right) \frac{1}{k_\perp} e^{ik_\parallel (\Delta x \cos \phi + \Delta y \sin \phi)} e^{ik_\perp |\Delta z|} \tag{37}
\]
The assisted decay rate, when atom A is in the state $|n^A\rangle$, can be derived with the same procedure of the resonant force: the rate is simply the imaginary part of the T matrix:

$$
\Gamma_n (r_A, r_B) = \frac{2\mu_0^2}{\hbar} \sum_{k < n} (\omega_{nk}^A)^4 \text{Im} \left[ d_{nk}^A \cdot G^{(0)} (r_A, r_B, \omega_{nk}^A) \cdot \alpha^B (\omega_{nk}^A) G^{(0)} (r_B, r_A, \omega_{nk}^A) \cdot d_{kn}^A \right].
$$

(38)

where we have assumed atom B is isotropic. Making use of Eq. (36), the assisted rate can be expressed as an angular integral of the emission rate density $\gamma_n$:

$$
\Gamma_n (r_A, r_B) = \int_0^{2\pi} d\varphi \int_0^\infty dk_|| \gamma_n (r_A, r_B, k_||)
$$

(39)

where:

$$
\gamma_n (r_A, r_B, k_||) = \frac{2\mu_0^2}{\hbar} \sum_{k < n} (\omega_{nk}^A)^4 \text{Im} \left[ d_{nk}^A \cdot G^{(0)} (r_A, r_B, \omega_{nk}^A) \cdot \alpha^B (\omega_{nk}^A) G^{(0)} (r_B, r_A, \omega_{nk}^A) \cdot d_{kn}^A \right]
$$

(40)

The recoil rate $R_n$, which describes the momentum carried away by an excitation with lateral momentum $\hbar k_||$, reads:

$$
R_n (r_A, r_B, \varphi) = \int_0^\infty dk_|| k_|| \gamma_n (r_A, r_B, k_||)
$$

(41)

Inserting the free-space Green’s tensor into (40), and simplifying under the assumption that the excited atom is an effectively two-level system, we find the following result for two atoms positioned on the $z$ axis at separation $r > 0$:

$$
R (r, \varphi) = -\frac{\alpha^B (\omega_\lambda) d_\lambda^2}{16\pi^3 r^3 \epsilon_0^2} \int_0^\infty dk_|| k_||^2 e^{i(r k_\perp + \xi)} \left[ 2r^2 k_\perp^2 (\xi + i) + r^2 k_\perp^2 (\xi^2 + 3i\xi - 3) k_|| \cos \varphi - \xi^2 (i\xi^2 + \xi + i) r^2 (i\xi^2 - \xi - i) k_||^2 \cos^2 \varphi \right].
$$

(42)

This integral is elementary, except for the terms containing a $k_\perp$ in the denominator. To calculate these we make use of the following:

$$
\int_0^\infty dk_|| k_||^2 e^{i k_\perp r} = \frac{\pi \xi}{2r^2} H_1 (\xi),
$$

$$
\int_0^\infty dk_|| k_||^4 e^{i k_\perp r} = \frac{3\pi \xi^2}{2r^4} H_2 (\xi)
$$

where $\xi = \omega_\lambda r/c$ and $H_n (x)$ are the Hankel functions of the first kind. After some algebra one finds that the emission spectrum is given by

$$
R (r, \varphi) = \frac{d_\lambda^2 \alpha^B (\omega_\lambda)}{64\pi^3 r^3 \epsilon_0^2} \left[ f_1 (\xi) + f_2 (\xi) \cos (2\varphi) + f_3 (\xi) \cos (\varphi) \right]
$$

(43)

where:

$$
f_1 (\xi) = \pi \xi^2 \left\{ 2\xi J_1 (\xi) \left[ (\xi^2 - 1) \cos \xi - \xi \sin \xi \right] + 3J_2 (\xi) \left[ 5\xi \sin \xi - (\xi^2 - 5) \cos \xi \right] - 2\xi Y_1 (\xi) \left[ (\xi^2 - 1) \sin \xi + \xi \cos \xi \right] + 3Y_2 (\xi) \left[ (\xi^2 - 5) \sin \xi + 5\xi \cos \xi \right] \right\},
$$

$$
f_2 (\xi) = 3\pi \xi^2 \left\{ J_2 (\xi) \left[ (1 - \xi^2) \cos \xi + \xi \sin \xi \right] + Y_2 (\xi) \left[ \xi \sin \xi + \cos \xi \right] - \sin 2\xi \right\},
$$

$$
f_3 (\xi) = 48\xi (\xi^2 - 3) \cos (2\xi) + 8 (9 - 15\xi^2 + 2\xi^4) \sin (2\xi)
$$

(44)

where $J_n (\xi)$ and $Y_n (\xi)$ are Bessel functions of the first and second kind, respectively. These are related to the Hankel functions via $H_n (x) = J_n (x) + i Y_n (x)$. The emission spectrum is asymmetric in the lateral direction $\hat{\xi}$:

$$
\int_{-\pi/2}^{\pi/2} d\varphi R (r, \varphi) - \int_{-\pi/2}^{\pi/2} d\varphi R (r, \varphi) = \frac{d_\lambda^2 \alpha^B (\omega_\lambda)}{16\pi^3 \epsilon_0^2 r^3} f_3 (\xi)
$$

(45)
In particular the lateral resonant force is a recoil force because of this asymmetric emission, see Eq. (8):

\[
F_{A,x}(r,t) = -p_1(t) \int_0^{2\pi} d\varphi \int_0^{\infty} dk_\parallel k_\parallel (\hbar k_\parallel \cos \varphi) \gamma (r,k_\parallel) = -p_1(t) \int_0^{2\pi} d\varphi R(r,\varphi) \cos \varphi. \tag{46}
\]