Quantum control of atoms at ultra-short distances from surfaces would open a new paradigm in quantum optics and offer a novel tool for the investigation of near-surface physics. Here, we investigate the motional states of atoms that are bound weakly to the surface of a hot optical nanofiber with optimized mechanical properties. We theoretically demonstrate that these states are quantized despite phonon-induced decoherence. We further show that it is possible to influence their properties with additional nanofiber-guided light fields and suggest heterodyne fluorescence spectroscopy to probe the spectrum of the quantized atomic motion. Extending the optical control of atoms to smaller atom-surface separations could create opportunities for quantum information processing and instigate the convergence of surface physics, quantum optics, and the physics of cold atoms.

Obtaining optical control over individual atoms close to surfaces would enable tremendous advances in fundamental research and offer significant opportunities for technological applications. For instance, trapping atoms closer to a waveguide increases their coupling to the evanescent tail of guided light fields. The increased emission into the waveguide aids the exploration of novel effects in quantum optics [1] and benefits powerful light-matter interfaces useful for quantum information processing [2]. Moreover, the measurement precision of effects in surface physics such as dispersion forces could profit from isotopically clean atomic probes with well-defined initial conditions and long interrogation times [3–5]. Precise control over the motional and electronic degrees of freedom of atoms near surfaces would, therefore, provide advantages for both quantum optics and surface physics and could ultimately enable the transfer of techniques between these two disparate fields. At present, cold atoms can be optically trapped at distances of a few hundred nanometers from surfaces [6–15]. Attempts to further reduce the atom-surface separation face the challenge that attractive dispersion forces start to dominate over optically or magnetically induced traps and can lead to adsorption [16]. Conversely, the omnipresence of dispersion forces has stimulated ideas to exploit them for trapping atoms in the first place [17–19]. In previous works on the optical control of adsorbed atoms [20–25], it remained unclear whether the motional states are quantized despite decoherence due to the interaction with the surface [26–28] and how to optimally probe and manipulate this system.

Here, we theoretically study individual atoms bound directly to the surface of an optical nanofiber [29–31]. We consider two cases: First, atoms adsorbed to the nanofiber, and second, surface-bound atoms in a hybrid potential created by adding an attractive optical force. We focus on weakly bound motional states with binding energies corresponding to a few megahertz (MHz) since these states can efficiently be probed with light. We account for the finite linewidth of transitions between motional states, which is caused by thermal vibrations (phonons) of the nanofiber. We identify a parameter regime in which the atomic motion normal to the surface is quantized both for adsorbed and for hybrid surface-bound atoms despite their interaction with nanofiber phonons. Interestingly,
the linewidths are limited by phonon-induced dephasing rather than state depopulation. We further show that the spectrum of the quantized atomic motion can be probed using heterodyne fluorescence spectroscopy, where a far-detuned probe laser drives transitions between motional states.

We consider cesium atoms bound to a silica nanofiber for the sake of concreteness. The existence of adsorbed states of cesium on silica is undisputed [32–34]. However, a quantization of the motion of the adatoms normal to the surface can only be observed if transitions between different motional states have linewidths that do not exceed the splitting between the transition frequencies in the absence of vibrations. The finite linewidths arise both from the depopulation and the dephasing of the involved motional states. The interaction with thermal phonons is the dominant mechanism causing depopulation both for adsorbed [26, 27] and optically trapped atoms [36], and leads to dephasing as well. We assume that the system is engineered in such a way that the relevant phonon modes are reflected at the tapered ends of the nanofiber region, creating a phonon cavity of length $L$. Such a cavity provides a degree of control over the nanofiber phonon modes and could, for instance, be realized by optimizing the nanofiber tapers [35]. To calculate the total linewidth of transitions between the motional states of an individual atom, we describe the coupled dynamics of the atomic motion and the nanofiber phonons using the Hamiltonian

$$\hat{H} = \hat{H}_{\text{ext}} + \hat{H}_{\text{phn}} + \hat{H}_{\text{ext-phn}}. \quad (1)$$

The atom Hamiltonian $\hat{H}_{\text{ext}} = \hat{p}^2/(2M) + V(\hat{r})$ describes the motion of the atom of mass $M$ in the cylindrically symmetric adiabatic potential $V(r)$. The operator $\hat{r}$ represents the distance of the atom from the axis of nanofiber and $\hat{p}$ the momentum of the atom. The term $\hat{H}_{\text{phn}}$ describes the dynamics of the nanofiber phonons, and the term $\hat{H}_{\text{ext-phn}}$ accounts for the atom-phonon coupling.

The potential $V(r)$ arises from both optical dipole forces [37, 38] and surface effects [28, 39]. We approximate the total potential as $V(r) = V_{\text{opt}}(r) + V_{\text{ad}}(r)$. Nonadiabatic corrections are only relevant for sufficiently strong light fields [40] but could potentially be studied in this setup. The optical potential $V_{\text{opt}}(r)$ can be controlled in experiments by tuning the laser beams that are coupled into the nanofiber. To create the hybrid light- and surface-induced potential, a circularly polarized, guided, running-wave light field with a frequency red detuned relative to the cesium $D_2$ line is injected into the nanofiber. The resulting potential can be calculated given the polarization of the atom and the wavelength, polarization, and power of the guided light fields [38, 41, 42]. Unlike in nanofiber-based two-color traps [10, 11], the potential is cylindrically symmetric and there is no repulsive optical force to prevent the atom from accessing the nanofiber surface. The potential $V_{\text{ad}}(r)$ is determined by the choice of atom species and nanofiber material. It is responsible for the adsorption (physisorption) of atoms on the nanofiber surface and is predominantly due to two effects: the Casimir-Polder interaction and the exchange interaction [16, 43, 44]. The attractive Casimir-Polder force (dispersion force) dominates over optical forces at atom-surface separations below a few tens of nanometers [38, 39]. The exchange interaction becomes relevant when electrons orbiting the atom begin to overlap with electrons in the nanofiber surface [16, 43, 45]. It causes a strong repulsion of the atom immediately at the nanofiber surface. We model the adsorption potential as

$$V_{\text{ad}}(r) = -C(r - R)^{-3} + D(r - R)^{-12}. \quad (2)$$

Here, $r$ is the radial distance of the atom from the nanofiber axis and $R$ is the radius of the nanofiber; see the inset in Fig. 1a. The first term in Eq. (2) is the (nonrevered) dispersion force between an atom and a half space, an approximation that is sufficient for our purpose [46]. The constant $C > 0$ can be calculated [47–49] and determined experimentally. For a cesium atom and a silica surface $C/h = 1.18$ THz nm$^3$ [50], where $h$ is Planck’s constant. The second term in Eq. (2) is a standard heuristic model for the exchange energy [45]. The constant $D > 0$ can be inferred from the minimum $V_{\text{min}}$ of the adsorption potential $V_{\text{ad}}(r)$. We use $V_{\text{min}}/h = -128$ THz [32, 33], which yields $D/h = 96.5$ kHz nm$^{-12}$. While there are alternative models for the exchange energy [45], the results presented in the following are qualitatively independent of the exact choice of model and its parameters [51].

In Fig. 1a, we plot the potential $V(r)$ as a function of the atom-surface separation. The hybrid light- and surface-induced potential is realized by launching into the nanofiber a light field with a free-space wavelength of 1064 nm and a power $P_t = 1$ mW. We also show the potential of a typical nanofiber-based two-color optical dipole trap for comparison; see the Supplemental Material for details [51]. We assume a relative permittivity of $\epsilon = 2.1$ [53] and a nanofiber radius of $R = 305$ nm (the largest radius compatible with the single-mode regime for the light fields of the two-color trap [54]).

The radial motional states have frequencies $\omega_\nu$ and wavefunctions $\psi_\nu(r) \equiv \sqrt{\nu} \langle r|\nu \rangle$ that are obtained by solving the time-independent Schrödinger equation

$$\left[ -\frac{\hbar^2}{2M} \partial_r^2 + V(r) \right] \psi_\nu(r) = \hbar \omega_\nu \psi_\nu(r). \quad (3)$$

Here, the index $\nu$ counts the motional quanta in radial direction. The motion in azimuthal and axial direction can be neglected [52], so $H_{\text{ext}} = \hbar \sum_\nu \omega_\nu |\nu \rangle \langle \nu |$. We solve Eq. (3) numerically [55]. In Fig. 2, we plot the spectrum $\omega_\nu$ and some example wavefunctions $\psi_\nu(r)$ using $M = 2.21 \times 10^{-25}$ kg [56]. Fig. 2a shows weakly bound states of an adsorbed atom with binding energies of a few MHz up to the dissociation limit. Fig. 2b shows surface-bound states in the hybrid light- and surface-induced potential. While the expected center-of-mass position of an atom in these states is on the order of 100 nm, there is no potential barrier to keep the atom from accessing the surface.
At second order, we only retain terms describing resonant interaction with flexural phonons and are resonant with phonon modes \( [36] \). Transitions between the atom states in color traps is dominated by their interaction with flexural modes of a nanofiber can be calculated analytically \([57]\); the corresponding bosonic ladder operators. The phonon modes have large decay rates \( \omega_{\nu} \ll |g_{\mu\nu}|, |G_{\mu\nu}| \) compared to the coupling rates, the phonon modes can be adiabatically eliminated to obtain an effective description of the atom motion in the presence of the thermal phonon bath \([52]\).

One can then show that if a transition \( \nu' \leftrightarrow \nu \) between different motional states is externally driven, its resonance has a finite phonon-induced linewidth (full width at half maximum) of

\[
\Gamma_{\nu'\nu} = \Gamma^{(1)}_{\nu'\nu} + \Gamma^{(2)}_{\nu'\nu},
\]

see \([52]\). Here, \( \Gamma^{(1)}_{\nu'\nu} = \Gamma_{\nu'\nu} + \Gamma_{\nu\nu}' \) is the broadening due to depopulation of the two motional states caused by phonon absorption and emission through \( \hat{h}^{(1)}_{\text{ext-phon}} \). The depopulation rate \( \Gamma_{\nu'\nu}^{d} \approx \Gamma_{\nu'\nu} + \Gamma_{\nu\nu}' \) of each state is dominated by transitions to the nearest neighboring states. It is beneficial to work with a small phonon cavity to minimize \( \Gamma_{\nu'\nu} \).

For our case study, we choose a cavity sufficiently small such that the frequency \( \omega_{\nu} = \pi^2 R \sqrt{E/\rho}/(2L^2) \) of the fundamental cavity mode \( \mu_1 \) is larger than the transition frequencies \( |\omega_{\nu+\nu'}| \) of interest. Here, \( E \) is the Young modulus of the nanofiber (\( E = 72.6 \text{ GPa} \) for fused silica \([53]\)). In this limit, \( \Gamma_{\nu}^\pm \) is determined by the nonresonant coupling to the fundamental mode. As result,

\[
\Gamma_{\nu}^\pm \approx 4\bar{n} \left| g_{\mu_1(\nu+\nu')} \right|^2 \frac{1}{Q} \omega_1.
\]

where \( \bar{n} \) is the thermal population and \( Q = \omega_1/\kappa_1 \) the quality factor of the fundamental cavity mode. In deriving Eq. (9), we assume \( \bar{n} \approx k_B T/\hbar \omega_1 \gg 1 \) where \( T \) is the temperature of the nanofiber and \( k_B \) is the Boltzmann constant. The second contribution in Eq. (8),

\[
\Gamma^{(2)}_{\nu'\nu} \approx 16\bar{n}^2 \left( g_{\mu\nu} \right)^2 \frac{1}{Q} \omega_1.
\]
work either at a sufficiently low nanofiber temperatures to ensure that the linewidths between the electronic ground and excited state of the atom is the position-dependent relative permittivity.

is primarily caused by dephasing between the motional states due to the resonant coupling to the fundamental mode through $\hat{H}^{(2)}_{\text{ext-pha}}$. Here, $G_{\mu \nu \nu'} \equiv (G_{\mu \nu \nu'} - G_{\mu \nu})/2$. To ensure that the linewidths $\Gamma_{\nu \nu'}$ are smaller than the separation between transition frequencies $\omega_{\nu \nu'}$, one can work either at a sufficiently low nanofiber temperatures $T$ or with a sufficiently small flexural cavity; see [52] for details. The former has the disadvantage of limiting the use of lasers due to heating by absorption in the nanofiber. Hence, we choose to operate at room temperature for our case study and use a cavity of length $L = 5\mu m$. We further assume that the fundamental cavity mode has a quality factor of $Q = 100$. In this case, the linewidth is limited by dephasing, that is, $\Gamma_{\nu \nu'}^{(2)} \gg \Gamma_{\nu \nu'}^{(1)}$, and transitions between the motional states shown in Fig. 2 can be resolved as we now argue.

We propose to measure the spectrum of the quantized nanofiber-bound states using heterodyne fluorescence spectroscopy, see Fig. 1b, which allows the observation of the quantized motion of atoms in optical potentials [58]. To this end, a cloud of laser-cooled atoms is prepared around the nanofiber. The nanofiber-bound states are occupied according to a dynamical equilibrium of atoms being adsorbed and dissociated. These processes are mediated by the nanofiber phonons [26, 27], and the resulting occupation of the bound levels depends on the temperature of the nanofiber. Light from a single laser with a frequency $\omega_p$ far detuned from the transition between the electronic ground and excited state of the atom is split into two beams; see Fig. 1b. The first beam is used to probe the atom; the second one serves as a local oscillator. The probe beam is coupled into the nanofiber with circular polarization. A guided probe photon can be scattered inelastically by a bound atom through the evanescent electric field, changing its frequency to $\omega_s$ and causing the atom to change its motional state from $\nu$ to $\nu'$ while remaining in its electronic ground state. This process is most likely if $\omega_s - \omega_p + \omega_{\nu \nu'} = 0$, creating sidebands in the spectrum of the probe beam. After transmission through the nanofiber, the probe beam is recombined with the local oscillator. The beat signal is detected with a photodetector. The frequency of the local oscillator is shifted by an offset $\Delta\omega$ to separate the Stokes and anti-Stokes sidebands. Its polarization is matched to that of the probe beam. This setup is only sensitive to the radial motion of bound atoms [52]. The power $P$ of the scattered light as a function of the difference $\omega \equiv \omega_s - \omega_p$ can be inferred from the spectrum of the photocurrent.

The spectroscopy can be modeled by the Hamiltonian

$$\hat{H}' = \hat{H} + \hat{H}_{\text{int}} + \hat{H}_{\text{pha}} + \hat{H}_{\text{int-pha}}$$

(11)

where $\hat{H}_{\text{pha}} = \hbar \sum_\eta \omega_\eta \hat{a}_\eta^\dagger \hat{a}_\eta$ describes the nanofiber-guided photon modes $\eta$ and $\hat{H}_{\text{int-pha}} = -\mathbf{d} \cdot \mathbf{E}(\hat{r})$ is the dipole coupling [52]. Here, $\mathbf{E}$ is the electric field and $\mathbf{d}$ is the dipole moment of a single atom. While the probe beam is far detuned, the electronic structure of the atom can be modeled as an effective two-level system with transition frequency $\omega_0$, excited state $|e\rangle$, and Hamiltonian $\hat{H}_{\text{int}} = \omega_0 |e\rangle \langle e|$. One can show that the power of scattered light as a function of the difference $\omega$ is approximately [52]

$$P(\omega) \propto \sum_{\nu, \nu' \neq \nu} \frac{\Gamma_{\nu \nu'}}{2} n(\nu) |\mathcal{F}_{\nu \nu'}|^2$$

(12)

Since the potential $V(\hat{r})$ is not harmonic, this spectrum contains a separate sideband of width $\Gamma_{\nu \nu'}$ at the frequency $\omega_{\nu \nu'}$ of each transition $\nu \leftrightarrow \nu'$ between radial motional states. The amplitude of each sideband is proportional to the occupation $n(\nu)$ of the state $\nu$ and the corresponding Franck-Condon factor

$$\mathcal{F}_{\nu \nu'} \equiv \frac{E_{\eta \eta'}}{\sqrt{2\pi}} \int_0^{\infty} \psi_{\nu'}^* (r) \mathcal{E}_{\eta \eta'} (r) \cdot \mathcal{E}_{\eta \eta'} (r) \psi_{\nu}(r) \, dr$$

(13)

where we define $E_{\eta} \equiv \sqrt{\hbar \epsilon_0 \omega_\eta}$. Here, $\epsilon_0$ is the vacuum permittivity, the index $\eta \eta'$ ($\eta$) comprises the quantum numbers of the nanofiber-guided probe (scattered) photon, and $\mathcal{E}_{\eta}(r)$ is the radial partial wave of the corresponding electric mode field of the fundamental HE11 mode of a nanofiber [54, 59, 60]. The mode fields are normalized according to $\epsilon_0 \int_0^{\infty} \epsilon(r) \mathcal{E}_{\eta}(r) \cdot \mathcal{E}_{\eta}(r) \, dr = 1$ where $\epsilon(r)$ is the position-dependent relative permittivity.

In Fig. 3a, we plot the anti-Stokes sidebands corresponding to downward transitions between the adsorbed
states shown in Fig. 2a, assuming a nanofiber temperature of $T = 300 \text{ K}$. The spectrum in Fig. 3b corresponds to the hybrid surface-bound states shown in Fig. 2b, assuming $T = 420 \text{ K}$ based on the power $P_r$ of the laser beam coupled into the nanofiber and the measurements reported in Ref. [61]. In both cases, transitions between neighboring levels are resolved. Examples of such transitions are indicated by the dashed lines. Transitions between levels that are further separated in $\nu$ appear as smaller, interstitial peaks. In plotting Fig. 3, we choose a wavelength of 1000 nm for the probe laser and approximate the occupation of all relevant states as equal since the frequency interval they cover is much smaller than $k_B T$. The signal decreases for larger $\omega$ since the involved states have a smaller spatial extent, resulting in lower Franck-Condon factors and a lower probability to scatter photons. For this reason, we focus on states with binding energies of a few MHz. The additional red-detuned light field increases the scattering probability in Fig. 2b by widening the wavefunctions: The resonances highlighted in Fig. 2a and Fig. 3b involve states with similar binding energies, but the signal is increased by about one order of magnitude in the latter case, lifting resonances above energies, but the signal is increased by about one order.

In summary, we analyze the spectrum and phonon-induced linewidths of the motional states of a cesium atom bound directly to the surface of an optical nanofiber. We distinguish two cases: adsorbed atoms in the absence of light, and atoms additionally bound by optical forces. We find that the phonon-induced linewidth of transitions between states with binding energies of a few MHz can be smaller than the spacing of the transitions, allowing to resolve quantized motional states. We further propose to probe these states using heterodyne fluorescence spectroscopy. The additional attractive light field enhances the expected signal compared to purely adsorbed atoms.

When working at room temperature, careful optimization of the nanofiber’s mechanical properties is required to resolve the quantization of the motional states, which could explain why it has not previously been observed in similar experiments. The proposed technique can be adapted for other waveguide geometries, including chip-based implementations, and is expected to work for other combinations of atom species and waveguide materials.

We thank Jürgen Volz and Carlos Gonzalez-Ballestero for helpful discussions. Financial support by the Austrian Academy of Sciences (ÖAW, ESQ Discovery Grant QuantSurf), the Studienstiftung des Deutschen Volkes, and the Alexander von Humboldt Foundation is gratefully acknowledged.

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Supplemental Material for ‘Probing Surface-Bound Atoms with Quantum Nanophotonics’

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In this supplement, we provide details on the calculation of the phonon-induced linewidths and the fluorescence spectra. In Sec. S1, we summarize the relevant phononic and photonic modes of the nanofiber. In Sec. S2, we discuss the motional states of adsorbed and surface-bound atoms shown in Fig. 2 of the paper. We describe how they couple to flexural cavity phonons and how to calculate the resulting finite linewidths of transitions between motional states. In Sec. S3, we discuss motional states of atoms in nanofiber-based two-color traps. We describe how they couple to traveling flexural phonons and how to calculate the resulting depopulation rates of motional states, both numerically and analytically in the limit of a harmonic trap potential. We use these results to verify our numerical calculations and as a benchmark for the power of the spectroscopy signal from surface-bound atoms. In Sec. S4, we derive the spectra of light scattered by nanofiber-bound atoms when probed with a nanofiber-guided light field. These spectra are shown in Fig. 3 of the paper.

S1. NANOFIBER MODES

It is useful to quantize both the displacement field $\hat{u}(r)$ and the electric field $\hat{E}(r)$ in terms of eigenmodes of the nanofiber, modeled as a cylinder of radius $R$.

A. Flexural Phonons

The thermal vibrations of a nanofiber can be described using linear elasticity theory. The dynamical quantity of linear elasticity theory is the displacement field $u(r,t)$ that indicates how far and in which direction each point $r$ of a body is displaced from its equilibrium position [1, 2]. Canonical quantization of linear elasticity theory in terms of a set of vibrational eigenmodes can be performed in the usual way [3]. The resulting displacement field operator in the Schrödinger picture is

$$\hat{u}(r) = \sum_{\mu} U_{\mu}(r) \hat{b}_{\mu} + \text{H.c.}$$  \hspace{1cm} (S1)

Here, $w_{\mu}(r)$ are the mode fields associated with the phonon modes, $\mu$ is a multiindex suitable for labeling the modes, $\hat{b}_{\mu}$ are the corresponding bosonic ladder operators, and H.c. indicates the Hermitian conjugate. The mode density is $U_{\mu} \equiv \sqrt{\hbar/2\rho \omega_{\mu}}$, where $\rho$ denotes the mass density of the nanofiber and $\omega_{\mu}$ are the phonon frequencies. The phonon Hamiltonian takes the form $\hat{H}_{\text{phn}} = \hbar \sum_{\mu} \omega_{\mu} \hat{b}_{\mu}^{\dagger} \hat{b}_{\mu}$. The eigenmodes of a nanofiber (modeled as a homogeneous, and isotropic cylinder) are well known [1, 4, 5]. In cylindrical coordinates $(r, \varphi, z)$, the mode fields factorize into partial waves

$$w_{\mu}(r) = W_{\mu}(r) e^{ij(\varphi + pz)} \quad \text{or} \quad w_{\mu}(r) = W_{\mu}(r) \frac{1}{\sqrt{\pi L}} e^{ij \varphi} \sin(pz),$$  \hspace{1cm} (S2)

where $p$ is the propagation constant along the nanofiber axis and $j \in \mathbb{Z}$. The left expression corresponds to the mode fields of an infinitely long nanofiber. It models traveling phonons on a long nanofiber that are not reflected at its tapered ends. In this case, $p \in \mathbb{R}$. The right expression models the standing waves of a finite nanofiber (a phonon cavity) located at $z \in [0, L]$ with fixed ends that reflect phonons. Such a cavity supports phonons with $p = \pi m/L$, where $m = 1, 2, \ldots$. Transitions between motional states in a nanofiber-based two-color trap are dominated by flexural phonon modes with $j = \pm 1$ [6]. The continuum of traveling flexural phonons can be labeled by $\mu = (p, j)$, and the discrete set of cavity modes by $\mu = (m, j)$. Flexural phonons with kHz to MHz frequencies that are relevant here have
wavelengths much larger than the radius of the nanofiber. In this limit, the radial partial waves \( \mathbf{W}_\mu(r) \) have vector components
\[
\mathbf{W}_\mu^r(r) = \frac{1}{R}, \quad \mathbf{W}_\mu^\varphi(r) = \frac{ij}{R}, \quad \mathbf{W}_\mu^z(r) = -\frac{ip}{R},
\]
which are normalized according to \( \int_0^R \mathbf{W}_\mu(r)^2 r \, dr = 1 \) leading order in \( pR \). These flexural modes form a single band in the \((\omega_\mu, p)\) plane with a dispersion relation \( \omega_\mu = cRp^2/2 \) that is quadratic in the low frequency limit \([6]\). In the case of a flexural mode cavity, the phonon spectrum is hence \( \omega_\mu = m^2\pi^2R\sqrt{E/\rho}/(2L^2) \). The effective speed of sound is \( v = \sqrt{E/\rho} \), where \( E \) is the Young modulus of the nanofiber material. For fused silica, \( E = 72.6 \text{ GPa} \) and \( \rho = 2.20 \text{ g/cm}^3 \) such that \( v = 5.74 \times 10^3 \text{ m/s} \) \([7]\).

### B. Nanofiber-guided Photons

In the paper, we propose to perform fluorescence spectroscopy of surface-bound states using a nanofiber-guided probe laser. We need to describe nanofiber-guided photons to model this spectroscopy scheme. The electromagnetic field in the presence of the nanofiber can be quantized based on the photonic eigenmodes of the system \([3, 8]\). The photonic eigenmodes of a nanofiber (modeled as a cylindrical step-index waveguide with relative permittivity \( \epsilon \)) are well known \([9, 10]\). The resulting Hamiltonian is \( \hat{H}_{\text{phot}} = \hbar \sum_{\eta} \omega_\eta \hat{\alpha}_\eta \hat{\alpha}^\dagger_\eta \), where \( \eta \) is a multi-index suitable for labeling the eigenmodes, \( \omega_\eta \) is the frequency of each eigenmode, and \( \hat{\alpha}_\eta \) is the corresponding bosonic ladder operator. The electric field operator in the Schrödinger picture is
\[
\hat{E}(r) = \sum_\eta E_\eta [\hat{\alpha}_\eta \mathbf{e}_\eta(r) + \text{H.c.}],
\]
where we define the mode density \( E_\eta \equiv \sqrt{\hbar \epsilon_0 \omega_\eta}/2 \) and \( \epsilon_0 \) is the vacuum permittivity. The electric field modes are of the form
\[
\mathbf{e}_\eta(r) = \frac{\mathbf{e}_\eta(r)}{2\pi} e^{i(m\varphi + kz)},
\]
with propagation constant \( k \in \mathbb{R} \) and azimuthal order \( m \in \mathbb{Z} \). These modes are quasi-circular polarized \([11]\). We are interested in photons in the single-mode regime of the nanofiber, that is, with frequencies below the cutoff frequency \( \omega_c \simeq 2.405 c/(R\sqrt{\epsilon - 1}) \) \([9]\). Here, \( c \) is the vacuum speed of light. For fused silica, \( \epsilon = 2.1 \) \([7]\) such that the silica nanofiber with a radius of \( R = 305 \text{ nm} \) considered in our case study has a cutoff frequency corresponding to a free-space wavelength of \( \lambda_c = 835.7 \text{ nm} \). In the single-mode regime, only modes on the HE\(_{11}\) band with azimuthal order \( m = \pm 1 \) are nanofiber-guided. For the setup considered in the paper, the fluorescence spectrum is independent of the sign of \( m \) and we may choose \( m = 1 \) without loss of generality. In this case, the radial partial waves of the electric mode field have vector components

\[
\begin{align*}
\mathbf{E}_\eta^r(r) &= \frac{\epsilon_\eta}{\alpha^2} \left[ k a J_1'(ar) - \frac{\omega_\eta}{c} \beta J_1(ar) \right], & \mathbf{E}_\eta^\varphi(r) &= -\alpha \frac{\epsilon_\eta}{b^2} \left[ k b K_1'(br) - \beta \omega_\eta \frac{K_1(br)}{r} \right], \\
\mathbf{E}_\eta^z(r) &= \alpha b J_1(ar), & \mathbf{E}_\eta^z(r) &= \alpha A_\eta K_1(br),
\end{align*}
\]

where \( a \equiv \sqrt{\omega_\eta^2/c^2 - k^2}, b \equiv \sqrt{k^2 - \omega_\eta^2/c^2} \) and \( v = c/\sqrt{\epsilon} \) is the speed of light inside the nanofiber. The functions \( J_m \) and \( K_m \) are Bessel functions and modified Bessel functions, respectively. The prime indicates the first derivative. We define
\[
\alpha \equiv \frac{J_1(aR)}{K_1(bR)}, \quad \beta \equiv \frac{(\epsilon - 1) k \omega_\eta}{Rc ab} \frac{J_1(aR)K_1(bR)}{aJ_1(aR)K_1'(bR) + bJ_1'(aR)K_1(bR)}. \tag{S7}
\]

The amplitude \( A_\eta \) is determined by the normalization condition \( \int_0^\infty re(r) \mathbf{E}_\eta^r(r) \cdot \mathbf{E}_\eta^r(r) r \, dr = 1 \). Here, \( e(r) \) is the relative permittivity as a function of the radial position. The dispersion relation \( \omega_\eta(k) \) is implicitly given by the frequency equation
\[
[aJ_1(aR)K_1'(bR) + bK_1(bR)J_1'(aR)] [aJ_1(aR)K_1'(bR) + c b K_1(bR)J_1'(aR)] = \left[ \frac{(\epsilon - 1) k \omega_\eta}{Rc ab} J_1(aR)K_1(bR) \right]^2. \tag{S8}
\]
The frequency equation has only one zero $\omega_\eta(k)$ in the single-mode regime.

## 2. Linewidths for Adsorbed and Surface-Bound Atoms

We provide details on the calculation of the motional states of adsorbed and surface-bound atoms shown in Fig. 2 of the paper. We also summarize how to calculate the linewidths of transition between the motional states due to the interaction with flexural cavity phonons. These linewidths are used to plot the spectra in Fig. 3 of the paper.

### A. Motional States

The potentials considered in the paper are cylindrically symmetric, that is, $V(\mathbf{r}) = V(r)$. The motional states $|\xi\rangle \equiv |\nu, l, q\rangle$ of an atom in these potentials, therefore, have wavefunctions of the form

$$\Psi_\xi(\mathbf{r}) = (\mathbf{r}|\nu, l, q\rangle = \frac{\psi_\nu(r)}{2\pi \sqrt{r}} e^{i(l\varphi + qz)}. \quad (S9)$$

The Hamiltonian describing the motion of the atom is $\hat{H}_{\text{ext}} = \hbar \sum_\xi \omega_\xi |\xi\rangle \langle \xi|$. The corresponding frequencies are $\omega_\xi = \omega_\nu + \hbar \eta^2/2M$ for an atom of mass $M$. Here, the quantum numbers $\nu \in \mathbb{N}$, $l \in \mathbb{Z}$, and $q \in \mathbb{R}$ label the excitations in radial, azimuthal, and axial direction, respectively. The radial partial waves $\psi_\nu(r)$ are obtained by solving the one-dimensional Schrödinger equation with the effective potential $V_l(r)$ $[12]$:

$$\left[ -\frac{\hbar^2}{2M} \partial_r^2 + V_l(r) \right] \psi_\nu(r) = \hbar \omega_\nu \psi_\nu(r), \quad V_l(r) \equiv V(r) + \frac{\hbar^2}{2M} \left( l^2 - \frac{1}{4} \right). \quad (S10)$$

The second term in the above potential is an angular momentum barrier. It can be neglected for azimuthal orders $l$ up to of a few hundred for adsorbed cesium atoms in weakly bound states considered in this paper. In that case, there is no coupling between the atomic motion in radial and azimuthal direction and $\psi_\nu(r) = \psi_r(r)$. Eq. (S10) then reduces to the Schrödinger equation

$$\left[ -\frac{\hbar^2}{2M} \partial_r^2 + V(r) \right] \psi_\nu(r) = \hbar \omega_\nu \psi_\nu(r) \quad (S11)$$

that we solve to calculate the states shown in the paper.

### B. Atom-Phonon Interaction

The coupling between atom motion and phonons arises because the phonons displace the potential, $V[\hat{\rho} - \hat{\tau}^R(\mathbf{R}, \varphi, z)]$. The interaction Hamiltonian is obtained by expanding the shifted potential to second order around $\mathbf{u} = \mathbf{0}$ and can be cast into the form $\hat{H}_{\text{ext-phon}} = \hat{H}_{\text{ext-phon}}^{(1)} + \hat{H}_{\text{ext-phon}}^{(2)}$ where

$$\hat{H}_{\text{ext-phon}}^{(1)} = \hbar \sum_{\mu \xi \xi'} \left( g_{\mu \xi \xi'} \hat{b}_\mu \hat{b}_{\mu}^\dagger |\xi\rangle \langle \xi| + \text{H.c.} \right),$$

$$\hat{H}_{\text{ext-phon}}^{(2)} = \hbar \sum_{\mu' \mu \xi \xi'} \left( \frac{K_{\mu' \mu \xi \xi'}}{2} \hat{b}_{\mu'} \hat{b}_{\mu} |\xi\rangle \langle \xi| + \text{H.c.} \right) + \hbar \sum_{\mu' \mu \xi \xi'} G_{\mu' \mu \xi \xi} \hat{b}_{\mu'} \hat{b}_{\mu} |\xi\rangle \langle \xi|. \quad (S12)$$

The coupling rates between atoms and cavity phonons are, at first order,

$$g_{\mu \xi \xi} = g_{\mu \nu \nu} \delta_{(l+j), l'} \left\{ \frac{1}{2} \{ \delta [q' - (q + p)] - \delta [q' - (q - p)] \} \right\}, \quad g_{\mu \nu \nu} = \frac{i}{\sqrt{2\pi}} \frac{A_{\nu \nu}}{\sqrt{\hbar \rho \omega_\nu LR}}, \quad (S13)$$

and, at second order,

$$K_{\mu' \mu \xi \xi} = G_{\mu' \mu \nu \nu} \delta_{l';(l+j)}[\delta], \quad G_{\mu' \mu \xi \xi} = G_{\mu' \mu \nu \nu} \delta_{l';(l+j)}[\delta], \quad G_{\mu' \mu \nu \nu} = \frac{1}{2\pi \rho \sqrt{\omega_\nu \omega_\mu LR^2}}, \quad (S14)$$
We use Pauli matrices \( \sigma^+ \) and \( \sigma^- \) are defined in the paper.

We focus on the radial motion of the atoms. Since phonons carry only little momentum, we neglect changes in the momentum of the atomic motion in the axial and azimuthal direction. To infer how the presence of thermal phonons affects the radial atomic motion, let us at first select two states \( |\nu_1\rangle \) and \( |\nu_2\rangle = |\nu_1 + 1\rangle \) that are neighbors in frequency. For the time being, we neglect all other atom states. The transition frequency \( \omega_0 \) can be neglected in our case study.

The effective Liouvillian is the rate \( \Gamma \) compared to the coupling rates, it is possible to obtain an effective description of the atom motion alone. If we further neglect nonresonant terms (i.e., terms that are not energy conserving) in the dissipator is \( \rho \), and the dissipator is \( \Gamma \).

In deriving Eq. (S16), we have redefined \( \hat{H}_{\text{ext}} \) to include a correction \( \Delta \omega_0 \equiv \sum \mu \bar{G}_\mu \bar{n}_\mu \) to the transition frequency \( \omega_0 \equiv \omega_{\nu_2} - \omega_{\nu_1} + \Delta \omega_0 \). The correction arises from \( \hat{H}_{\text{ext}}^{(2)} \) to the finite thermal population of the phonon bath. It can be neglected for the parameters used in the case study in the paper. We also neglect nonresonant terms (i.e., terms that are not energy conserving) in \( \hat{H}_{\text{ext}}^{(2)} \), since all phonon scattering, absorption, and emission processes are dominated by resonant terms. At this point, there are still terms proportional to \( \delta^+ \) and \( \delta^- \) remaining, which lead to transitions between the two atom states through two-phonon absorption, emission, or inelastic scattering at first order in \( \hat{H}_{\text{ext}}^{(2)} \). These processes contribute to the broadening of the resonance when the transition \( \nu_1 \leftrightarrow \nu_2 \) is externally driven. However, the coupling constants are much smaller than for the elastic two-phonon scattering processes generated by the terms \( \tilde{b}_\mu \tilde{b}_\mu \hat{\sigma}^2 \), which cause dephasing. As a result, the linewidth induced by \( \hat{H}_{\text{ext}}^{(2)} \) is dominated by dephasing due to the resonant \( \hat{\sigma}^2 \) terms retained in Eq. (S16).

### C. Effective Evolution of the Atomic Motion

In practice, the phonon modes have a thermal population and nonzero decay rates \( \kappa_\mu \) due to internal losses and their interaction with the environment (e.g., through the absorption of guided laser light and the clamping of the nanofiber). We model the dynamics of the joint atom-phonon state operator \( \hat{\rho} \) using the Liouvillian \( \mathcal{L} = \mathcal{L}_{\text{ext}} + \mathcal{L}_{\text{phn}} + \mathcal{L}_{\text{ext-phn}} \), where

\[
\mathcal{L}_{\text{ext}} \hat{\rho} = -\frac{i}{\hbar} [\hat{H}_{\text{ext}}, \hat{\rho}], \quad \mathcal{L}_{\text{phn}} \hat{\rho} = -\frac{i}{\hbar} [\hat{H}_{\text{phn}}, \hat{\rho}] + \sum _\mu \kappa_\mu (\tilde{n}_\mu + 1) D_{b_\mu} \hat{\rho} + \kappa_\mu \tilde{n}_\mu D_{b_\mu} ^\dagger \hat{\rho}, \quad \mathcal{L}_{\text{ext-phn}} \hat{\rho} = -\frac{i}{\hbar} [\hat{H}_{\text{ext-phn}}, \hat{\rho}],
\]

and the dissipator is \( D_{b_\mu} \hat{\rho} = \tilde{b}_\mu \hat{\rho} \tilde{b}_\mu ^\dagger - \{ \tilde{b}_\mu ^\dagger \tilde{b}_\mu , \hat{\rho} \} / 2 \). The steady-state of the phonon bath according to \( \mathcal{L}_{\text{phn}} \) is the thermal state \( \hat{\sigma}_\mu = e^{-\hat{H}_{\text{phn}}/(K_B T)} / \text{tr}[e^{-\hat{H}_{\text{phn}}/(K_B T)}] \) with thermal populations \( \tilde{n}_\mu \) determined by the Bose-Einstein distribution. Here, \( T \) is the temperature of the nanofiber. Since the transition frequency \( \omega_0 \gg |g_\mu|, |G_\mu| \) is large compared to the coupling rates, it is possible to obtain an effective description of the atom motion alone. If we further assume \( \kappa_\mu \gg |g_\mu|, |G_\mu| \), we can use adiabatic elimination to trace out the phonon modes [13, 14]. The dynamics of the state operator \( \hat{\mu} \) is then described by the Liouville–von Neumann equation \( \hat{\delta} \hat{\mu}(t) = \mathcal{L}_{\text{eff}} \hat{\mu}(t) \) with the effective Liouvillian

\[
\mathcal{L}_{\text{eff}} \hat{\mu} = -\frac{i}{\hbar} \left[ \hat{H}_{\text{eff}}, \hat{\mu} \right] + \Gamma^- D_{\hat{\sigma}^-} \hat{\mu} + \Gamma^+ D_{\hat{\sigma}^+} \hat{\mu} + \Gamma^z D_{\hat{\sigma}^z} \hat{\mu}, \quad \hat{H}_{\text{eff}} = \hbar \frac{\omega_{\text{eff}}}{2} \hat{\sigma}^z.
\]

Here, \( \Gamma^+ \) and \( \Gamma^- \) are the phonon-induced depopulation rates of the states \( \nu_1 \) and \( \nu_2 \), respectively, and \( \Gamma^z \) is the rate of phonon-induced depopulation between the two states:

\[
\Gamma^+ = 2 \sum _\mu |g_\mu|^2 \text{Re} \left[ \tilde{n}_\mu K^-_\mu + (\tilde{n}_\mu + 1) K^+_\mu \right], \quad \Gamma^- = 2 \sum _\mu |g_\mu|^2 \text{Re} \left[ (\tilde{n}_\mu + 1) K^-_\mu + \bar{n}_\mu K^+_\mu \right], \quad \Gamma^z = 2 \sum _\mu \tilde{n}_\mu (\tilde{n}_\mu + 1) G^2_\mu / \kappa_\mu,
\]

where

\[
K^\pm_\mu \equiv \frac{\kappa_\mu / 2}{(\kappa_\mu / 2)^2 + (\omega_0 \pm \omega_\mu)^2} + i \frac{\omega_0 \pm \omega_\mu}{(\kappa_\mu / 2)^2 + (\omega_0 \pm \omega_\mu)^2}.
\]

The transition frequency \( \omega_{\text{eff}} \equiv \omega_0 + \Delta_L \) is subject to the Lamb shift \( \Delta_L \equiv \sum _\mu (2\tilde{n}_\mu + 1)|g_\mu|^2 \text{Im} \left[ K^-_\mu + K^+_\mu \right] \), which can be neglected in our case study.
D. Linewidth of Transitions

To determine the phonon-induced linewidth of the transition \( \nu_1 \leftrightarrow \nu_2 \), we can, for instance, add a driving term \( \hat{H}_d(t) = \hbar \Omega [\hat{\sigma}^- e^{i \omega_d t} + \text{H.c.}] / 2 \) to Eq. (S18). In the limit of a driving that is weak compared the influence of the bath, \( \Omega \ll (\Gamma^+ , \Gamma^-) \), the steady-state population of the state \( |\nu_2\rangle \) is

\[
\langle \nu_2 | \hat{n}_\mu | \nu_2 \rangle \simeq \frac{\Omega^2}{2(\Gamma^- + \Gamma^+)} \frac{\Gamma/2}{\Delta^2 + (\Gamma/2)^2} + \frac{\Gamma^+}{\Gamma^- + \Gamma^+},
\]

(S21)

where \( \Delta \equiv \omega_d - \omega_\text{eff} \) is the detuning of the drive. The resonance in the population as a function of the detuning has a Lorentzian shape with linewidth (full width at half maximum) of

\[
\Gamma = \Gamma^- + \Gamma^+ + 4\Gamma^z.
\]

(S22)

The linewidth has two distinct contributions: \( \Gamma^{(1)} \equiv \Gamma^- + \Gamma^+ \) due to the depopulation of the two involved states, and \( \Gamma^{(2)} \equiv 4\Gamma^z \) due to the dephasing of the two states. By construction of the model Eq. (S16), we neglect depopulation induced by \( \hat{H}^{(2)}_{\text{ext-phon}} \) since it leads to a broadening that is smaller than \( \Gamma^{(2)} \).

It is straightforward to generalize to transitions between any of the radial motional states \( |\nu\rangle \). In analogy to Eq. (S22), we model the linewidth of the transition \( \nu \leftrightarrow \nu' \) between any two states as

\[
\Gamma_{\nu'\nu}^{(1)} \equiv \Gamma_{\nu'\nu}^{(1)} + \Gamma_{\nu'\nu}^{(2)}.
\]

(S23)

Here,

\[
\Gamma_{\nu'\nu}^{(2)} \equiv 8 \sum_{\mu} \bar{n}_\mu^2 \frac{G_{\mu\nu'\nu}}{\kappa_\mu}, \quad G_{\mu\nu'\nu} \equiv \frac{1}{4\pi} \frac{\mathcal{A}_{\nu'\nu}^{(2)} - \mathcal{A}_{\nu'\nu}^{(2)} B_{\nu'\nu}}{\rho \omega_\mu LR^2}
\]

(S24)
in analogy to Eq. (S20). Note that \( G_{\mu\nu'\nu} \in \mathbb{R} \). The rate \( \Gamma_{\nu'\nu}^{(2)} \) is dominated by the fundamental cavity mode \( \mu_1 \), since the coupling rates drop as \( \omega_\mu^{-2} \) with the phonon frequency. Hence,

\[
\Gamma_{\nu'\nu}^{(2)} \simeq 16\bar{n}^2 G_{\mu_1\nu'\nu}^2 \omega_1 Q = \frac{32}{\pi^2} \frac{k_B T^2 L^8 Q}{\hbar^2 R^9} \sqrt{\frac{\rho}{E^5}} \left[ \mathcal{A}_{\nu'\nu}^{(2)} - \mathcal{A}_{\nu'\nu}^{(2)} \right]^2,
\]

(S25)

where \( \bar{n} \) is the thermal population, \( \omega_1 \) the frequency, and \( Q = \omega_1/\kappa_1 \) the quality factor of the fundamental cavity mode.

The broadening \( \Gamma_{\nu'\nu}^{(1)} \) is the sum of the depopulation rates of both states. In general, transitions to any other state contribute to the depopulation rates. In the limit of large thermal populations \( \bar{n}_\mu \gg 1 \), we obtain

\[
\Gamma_{\nu'\nu}^{(1)} \equiv \Gamma_{\nu'\nu}^d + \Gamma_{\nu'\nu}^d = 2 \sum_{\nu' \neq \nu} \sum_{\mu} \bar{n}_\mu |g_{\mu\nu'\nu}|^2 \text{Re} \left[ K_{\mu\nu'\nu}^+ + K_{\mu\nu'\nu}^- \right], \quad \text{Re} K_{\mu\nu'\nu}^\pm = \frac{\kappa_\mu/2}{(\kappa_\mu/2)^2 + (|\omega_{\nu'\nu}|^2 + \omega_\mu)^2}
\]

(S26)
in analogy to Eqs. (S19) and (S20). Here, \( \omega_{\nu'\nu} \equiv \omega_{\nu'\nu} - \omega_\nu \) is the transition frequency and \( g_{\mu\nu'\nu} \) is defined in Eq. (S13). The state overlaps \( \mathcal{A}_{\nu'\nu}^{(1)} \) quickly decay with increasing distance \( |\nu' - \nu| \). As a result, it is often sufficient to include transitions to the states \( \nu'' = \nu \pm 1 \) closest in frequency when calculating \( \Gamma_{\nu'\nu}^d \). If the cavity is sufficiently small such that the fundamental cavity mode has a frequency \( \omega_1 \) larger than the relevant transition frequencies, \( \Gamma_{\nu'\nu}^{(1)} \) is dominated by the fundamental mode and we can approximate

\[
\Gamma_{\nu'\nu}^{(1)} \simeq \Gamma_{\nu'\nu}^d + \Gamma_{\nu'\nu}^d + \Gamma_{\nu'\nu}^d + \Gamma_{\nu'\nu}^d = 4\bar{n} |g_{\mu_1(\nu\pm1)\nu}|^2 \frac{1}{\omega_1 Q} = 16 \frac{k_B T L^5}{\pi^2 \hbar^2 R^9} \sqrt{\frac{\rho}{E^5}} |A_{(\nu\pm1)\nu}^{(1)}|^2,
\]

(S27)

which corresponds to Eq. (9) in the paper. We use Eqs. (S25) and (S26) to calculate the linewidths that appear in Fig. 3 of the paper, with relevant contributions only stemming from \( \Gamma_{\nu'\nu}^{(2)} \).

In the heterodyne fluorescence spectroscopy scheme we propose in the paper, transitions between all motional states are driven simultaneously. Transitions between states \( \nu \) and \( \nu' = \nu + 1 \) that are nearest neighbors in frequency are most likely and lead to resonances of the largest power, see Fig. 3 in the paper. Therefore, it is useful to focus on nearest-neighbor transitions to determine for which parameters the motional quantization can be resolved. For nearest-neighbor transitions, Eq. (S26) simplifies to

\[
\Gamma_{(\nu+1)\nu}^{(1)} \simeq 16 \sum_{m=1}^{\infty} \bar{n}_\mu |g_{\mu(\nu+1)\nu}|^2 \text{Re} \left[ K_{\mu(\nu+1)\nu}^- + K_{\mu(\nu+1)\nu}^+ \right].
\]

(S28)
wavefunctions in Fig. S2. To leading order in the phonon degrees of freedom, these states couple to flexural phonons.

In the regime $\nu' \gg 1$, the contribution $\Gamma_{\nu'\nu}$ can be neglected compared to $\Gamma_{\nu'\nu}$. Note that, for simplicity, we assume a constant quality factor $Q = \omega_{\mu}/\kappa_{\mu} = 100$ for all modes (in particular the fundamental mode decisive for the linewidth). This assumption cannot hold for arbitrarily large cavities: It is to be expected that the quality factor is reduced for modes with longer wavelengths, which in turn lowers $\Gamma_{\nu'\nu}/\Delta\omega$ compared to a simple extrapolation of Fig. S1.

In deriving Eq. (S28), we approximate the upward and downward depopulation rates of each state as equal. In this case, Eq. (S27) further simplifies to

$$
\Gamma_{(\nu+1)\nu}^{(1)} \approx 16n \frac{|g_{\mu\nu'\nu}|^2}{\omega_1} Q \frac{1}{Q} \frac{64 k_B TL^5}{\pi^2 R^3} \sqrt{\frac{\rho}{E^3}} |A_{\nu'\nu}^{(1)}|^2.
$$

In Fig. S1, we plot the contributions $\Gamma_{\nu'\nu}^{(1)}$ and $\Gamma_{\nu'\nu}^{(2)}$ to the linewidth as a function of the cavity length $L$ using Eqs. (S28) and (S29). We select the transition between the states $\nu = 261$ and $\nu' = 262$ shown in Fig. 2b of the paper. Below the horizontal dashed line, the linewidth $\Gamma_{\nu'\nu}$ is smaller than the separation $\Delta\omega$ to the next nearest-neighbor transition. In the regime $\Gamma_{\nu'\nu}/\Delta\omega \ll 1$, transitions between motional states can be resolved. This regime can be realized either by choosing a sufficiently small cavity, or by working at sufficiently low nanofiber temperatures. For the parameters chosen in Fig. S1, the contribution $\Gamma_{\nu'\nu}^{(1)}$ can be neglected compared to $\Gamma_{\nu'\nu}^{(2)}$. Note that, for simplicity, we assume a constant quality factor $Q = \omega_{\mu}/\kappa_{\mu} = 100$ for all modes (in particular the fundamental mode decisive for the linewidth). This assumption cannot hold for arbitrarily large cavities: It is to be expected that the quality factor is reduced for modes with longer wavelengths, which in turn lowers $\Gamma_{\nu'\nu}^{(2)}$ compared to a simple extrapolation of Fig. S1.

S3. LINEWIDTHS FOR OPTICALLY TRAPPED ATOMS

We derive the phonon-induced depopulation rate of radial motional states of atoms that are trapped in a two-color trap and interact with the traveling flexural phonons of a long nanofiber. This model is able to explain the heating rates observed in existing nanofiber-based atom trap setups [6]. We calculate the depopulation rates using the numerical methods also applied to the adsorbed and surface-bound states. We use these results to verify our numerical calculations by comparing them with analytical results obtained in the limit of a harmonic trap.

Fig. 1 of the paper shows a typical two-color trap potential. It is realized by launching two counterpropagating beams with a free-space wavelength of 1064 nm (red detuned with respect to the cesium $D_2$ line) and a combined power of $2 \times 2$ mW into the nanofiber, as well as a running-wave light field with a wavelength of 840 nm (blue detuned) and a power of 4.5 mW. All beams are linearly polarized, with a $\pi/2$ angle between the polarization planes of the blue- and red-detuned light fields. All other parameters are as in the case study presented in the paper. The trap minima are located in the polarization plane of the red-detuned light field. Close to the ground state of the trap, the radial motion of the atom decouples from its motion in the axial and azimuthal direction.

The radial motional states $|\nu\rangle$ can be obtained by solving Eq. (S11). We plot two examples of the corresponding wavefunctions in Fig. S2. To leading order in the phonon degrees of freedom, these states couple to flexural phonons.
We use this expression to verify our numerical methods: The numerical result obtained using Eq. (S31) and presented in Fig. S2 agrees well with the rate $\Gamma^{d}_\nu = 216$ Hz obtained analytically using Eq. (S34). These results are compatible with experimentally observed linewidths [6, 15, 16].
In the paper, we propose heterodyne fluorescence spectroscopy to probe the quantized spectrum of surface-bound motional states. Under suitable conditions [17], the resulting signal reveals Raman-type transitions between different states of the radial center-of-mass motion of atoms in their electronic ground state. This approach has advantages compared to the transmission [18, 19] or fluorescence excitation spectroscopy [20] used in previous experimental studies of surface-induced effects on atoms near optical nanofibers. These latter techniques probe surface-induced shifts between the ground state and a given excited electronic state of the atoms. In consequence, their resolution is limited by the natural linewidth of the excited electronic state. For the Raman spectroscopy technique proposed here, the surface-induced shifts only change the overall strength of the signal but not its shape. In consequence, the Raman spectroscopy is not limited by spectral width of the optically excited state and can provide access to the closely spaced energy levels shown in Fig. 2 of the paper.

To probe the radial motional states of atoms bound directly to the nanofiber surface, a circularly polarized probe laser with a frequency \( \omega_p \) detuned from resonance with the atom is coupled into the fiber as a traveling wave. The resulting polarization in the nanofiber region is quasi-circularly polarized, with azimuthal order \( m = \pm 1 \); see Sec. S1 B. The probe beam has a wavelength in the single-mode regime of the nanofiber, such that probe photons are guided on the HE\(_{11} \) band in the nanofiber region. We assume that the probe laser is sufficiently far detuned from resonance with transitions between the 6S and 6P manifolds of the cesium atom to treat the atom as an effective two-level system with ground state \( |g\rangle \), excited state \( |e\rangle \), and transition frequency \( \omega_0 \). Those photons that are scattered by the atom back into the nanofiber in the forward direction are recombined with the local oscillator on a beam splitter. The frequency \( \omega_s \) of a scattered photon is changed to \( \omega_s \) when the atom simultaneously changes its motional state, leading to motional sidebands in the spectrum of the probe beam. The frequency difference between the probe beam and the local oscillator results in a beat that can be observed with a photodetector. The local oscillator is shifted at an offset \( \Delta \nu \) such that the spectrum of the photocurrent contains sidebands at \( \omega_p + \Delta \nu \). This shift separates the Stokes- and anti-Stokes sidebands in the final signal and to choose the optimal working point for the photodetector. Moreover, the polarization of the local oscillator is matched to the polarization of the probe beam. In consequence, the beat signal is predominantly due to photons that are scattered without changing their polarization. This specific choice of polarizations eliminates the contribution of changes of the atoms’ azimuthal motional state to the spectroscopy signal, while the detection of light scattered in the forward direction minimizes the recoil in the axial motion of the atoms. As a result, the proposed spectroscopy configuration is only sensitive to the radial motion of the atoms, and the motional sidebands correspond to transitions \( \nu \rightarrow \nu' \) between different radial motional states.

The atom-phonon-photon system can then be described by the Hamiltonian \( \hat{H}' = \hat{H}_{\text{ext}} + \hat{H}_{\text{phn}} + \hat{H}_{\text{ext-phn}} + \hat{H}_{\text{int}} + \hat{H}_{\text{ph}} + \hat{H}_{\text{int-ph}} \) where the electronic structure of the atom is governed by \( \hat{H}_{\text{int}} = \hat{h} \omega_0 |e\rangle\langle e| \) and the atom interacts with the electric field through the dipole coupling \( \hat{H}_{\text{int-ph}} = -\hat{d} \cdot \hat{E}(\hat{r}) \). Here, \( \hat{d} \) is the dipole moment of the atom. This model assumes that the probe laser is weak such that multiple scattering of a photon by several atoms can be neglected, and it is sufficient to treat every atom individually. To predict the spectral distribution of the power \( P(\omega) \) of the scattered light as a function of the frequency difference \( \omega \equiv \omega_s - \omega_p \), one can calculate the steady-state of the system in the presence of a coherently driven laser mode and a thermal nanofiber phonon bath using a master equation approach [17, 21]. There is, however, an alternative way to approximate the resulting spectrum that is sufficient for the purpose of this paper: The motional states we consider have lifetimes corresponding to 2\( \pi / \Gamma_0 \) \( \sim 1 \) ms that are much longer than the time of 2\( \pi / \Gamma_0 \) \( \sim 100 \) ns it takes a probe photon to be absorbed and re-emitted by the atom. Here, \( \Gamma_0 \) is the lifetime of states in the 6P manifold of cesium. We can, therefore, treat the motional states as eigenstates for the duration of the scattering process and neglect their coupling to the nanofiber phonons. This approximation allows us to employ scattering theory to obtain the position and relative weight of the motional sidebands in the spectrum \( P(\omega) \).

In a second step, we then account for the finite linewidth of transitions between the motional states.

We assume that the probe laser has a sufficiently low power such that the atom only interacts with one photon at a time. The relevant transitions are, therefore, between states where the atom starts in its internal ground state \( |g\rangle \) and the motional state \( |\xi\rangle = |\nu, l, q\rangle \), and ends again in its ground state but with a different motional state \( |\xi'\rangle = |\nu', l', q'\rangle \). Simultaneously, a photon is scattered from the mode \( \eta_p \) to the mode \( \eta_s \). Since we detect only scattered photons that are still nanofiber-guided, propagate in the same direction, and have the same polarization, the modes \( \eta_p \) and \( \eta_s \) can only differ in their frequencies. Conservation of angular momentum then implies that \( m = \pm 1 \). Moreover, we can neglect the change in kinetic energy of the atom due to recoil along the nanofiber axis, so \( q' \approx q \). Energy conservation hence requires the detected photon to have a frequency shifted by \( \omega = \omega_s - \omega_p \). One can show using the resolvent [22] that the scattering matrix element for transitions \( \nu \rightarrow \nu' \) while changing the frequency of the photon by \( \omega \) is

\[
S_{\nu \nu'}(\omega) \equiv \frac{2\pi i}{\hbar^2} \frac{d}{\delta \left( \omega_{\nu \nu'} - \omega \right)} \frac{(d/3)^2 F_{\nu \nu'}}{\Delta + i \Gamma_0/2}.
\]

Here, \( \omega_{\nu \nu'} = \omega_{\nu'} - \omega_{\nu} \) is the frequency difference between the initial and the final radial motional state of the atom.
The motional states considered in the paper fall into a frequency interval that is small compared to the depth of the weakly bound states considered in the paper if the probe laser is detuned by a few MHz. We use Eq. (S31) to calculate the linewidths, assuming that the linewidths of atoms trapped in a surface-induced potential. In Fig. S3a, we plot the spectrum due to transitions between the optically trapped states shown in Fig. S2. The indicated transitions involve the states \( \nu = (253, 249) \), which have frequencies \( \omega_\nu = -2\pi \times (8.9, 20) \) MHz and lie deeper than the states shown in Fig. 2a of the paper.

and \( \Delta = \omega_p - \omega_0 \) is the detuning of the probe laser from resonance with the atom. Note that \( \omega_0 \) and \( \Gamma_0 \) are modified by the presence of the nanofiber compared to a cesium atom in free space. They depend on the distance between the atom and the nanofiber and hence on the radial motional state \( \nu \). In the following, we assume that differences in the transition frequency and decay rate can be neglected over the limited range of motional states we consider. The relative weights of the sidebands in Eq. (S35) are determined by the Franck-Condon factors

\[
\mathcal{F}_{\nu'\nu} = \frac{E_{\nu} E_{\nu'}}{(2\pi)^2} \int_0^\infty \psi^*_\nu(r) \mathcal{E}^\nu(r) \cdot \mathcal{E}^\nu_{\nu'}(r) \psi_{\nu'}(r) \, dr. \tag{S36}
\]

In deriving Eq. (S35), we (i) exploit that the scattering of a probe photon by the atom is sufficiently fast such that the motional state of the atom does not decay in the meantime; (ii) assume that \( |\Delta| \gg |\omega_{\nu'\nu}| \), which is the case for the weakly bound states considered in the paper if the probe laser is detuned by a few MHz; (iii) assume that the detuning is sufficiently large for the response of the atom to be isotropic, that is, \( (\mathbf{d} \cdot \mathbf{d}') = (d/3)^2 \delta^{ij} \) where \( d \in \mathbb{R} \) and \( \mathbf{d}' \) are components of the dipole moment \( d \) of the atom.

The power of the scattered light is \( P(\omega) \propto \sum_{\nu, \nu' \neq \nu} n(\nu) |S_{\nu'\nu}(\omega)|^2 \) where \( n(\nu) \) is the number of atoms initially in the motional state \( \nu \). In practice, the sharp sidebands in Eq. (S35) are broadened due to sources of noise and decoherence affecting either the laser or the motion of the atom. If the same laser source is used for both the probe beam and the reference beam, the frequency drift of the laser has no effect and the linewidths of the sidebands are determined by the decoherence of the motional atomic states. We can model the phonon-induced linewidths of the motional states by replacing the sharp sidebands in Eq. (S35) with Lorentzian resonances of the appropriate width \( \Gamma_{\nu'\nu} \) and the same total power:

\[
\delta (\omega_{\nu'\nu} - \omega) \rightarrow \frac{1}{\pi} \frac{\Gamma_{\nu'\nu}/2}{(\omega_{\nu'\nu} - \omega)^2 + (\Gamma_{\nu'\nu}/2)^2}. \tag{S37}
\]

The motional states considered in the paper fall into a frequency interval that is small compared to the depth of the potential \( V(r) \). In consequence, we can approximate the occupation \( n(\nu) \) of these states as constant. The power of the light scattered by the atom is therefore

\[
P(\omega) \propto \sum_{\nu, \nu' \neq \nu} \frac{\Gamma_{\nu'\nu}/2}{(\omega_{\nu'\nu} - \omega)^2 + (\Gamma_{\nu'\nu}/2)^2} |\mathcal{F}_{\nu'\nu}|^2 \tag{S38}
\]

as a function of the frequency difference between probe photons and detected photons.

In Fig. 3 of the paper, we show fluorescence spectra for adsorbed atoms and atoms in the hybrid light- and surface-induced potential. In Fig. S3a, we plot the spectrum due to transitions between the optically trapped states shown in Fig. S2. We use Eq. (S31) to calculate the linewidths, assuming that the linewidths of atoms trapped in...
two-color traps around a long nanofiber are limited by depopulation. We further approximate the population of the motional states as equal. In practice, the spectrum features additional sidebands from the motion in axial and azimuthal direction since the two-color trap confines the atom in all three spatial directions. These sidebands are omitted in Fig. S3. We use the power \( P_0 \) of the sideband corresponding to transitions between the radial ground state \( \nu = 0 \) and first excited state \( \nu = 1 \) as a reference and plot all spectra in units of \( P_0 \).

Fig. S3b shows the fluorescence spectrum for adsorbed atoms in a larger frequency interval than in Fig. 3a in the paper, involving states with larger binding energies. The corresponding wave functions have a much smaller spatial extent, which results in smaller Franck-Condon factors. Atoms in these states are, therefore, much less likely to scatter a nanofiber-guided photon and are more difficult to probe. Moreover, transitions with larger frequencies can no longer be resolved due to their increasing linewidths. For these reasons, we focus on states with binding energies of a few MHz and transition frequencies of a few hundred kHz in the paper.

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