

Nonlocal spatial correlations in an interacting atomic gas

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We report *in situ* measurements of the nonlocal two-body spatial correlation function $g^{(2)}(R)$ in an ultracold Bose gas with strong interactions using photoassociation of ultra-long-range Rydberg molecules (ULRRMs) as a spatially selective probe. This technique provides access to $g^{(2)}(R)$ on length scales from 60 to 180 nm, bridging the gap between short-range and long-range correlation measurements. The large s -wave scattering length of ^{86}Sr ($a = 811 a_0$) enables exploration of the universal regime where $g^{(2)}(R)$ is governed solely by two-body interactions. Comparison with a weakly interacting isotope, ^{84}Sr , highlights interaction-induced suppression of pair correlations. The measured correlation functions agree quantitatively with *ab initio* calculations based on the two-body reduced density matrix, without adjustable parameters. Our results establish Rydberg molecular photoassociation as a powerful tool for probing nonlocal correlations *in situ* in strongly interacting quantum gases.

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I. INTRODUCTION

Spatial correlation functions [1] herald distinctive properties of quantum many-body systems, such as ordering in quantum spin models and magnetism [2–5], fermionization in strongly interacting Bose gases in one dimension [6–10], and the absence of bunching in a Bose-Einstein condensate, which indicates the coherent state [11,12]. Powerful *in situ* probes have been developed to access short-range or local correlations in quantum gases, such as three-body recombination [7,9,11] and photoassociation [8] of dipole-dipole or van der Waals molecules [13]. Atomic gas microscopes [4,14–17] provide access to nonlocal spatial correlations at length scales on the order of a wavelength of light or larger. However, *in situ* probes in an intermediate regime have proven elusive.

A new *in situ* probe of the nonlocal, two-body spatial correlation function $g^{(2)}(R)$ in the intermediate regime utilizing photoassociation rates of ultra-long-range Rydberg molecules (ULRRMs) [18,19] was reported in Refs. [20,21] and used to observe bunching and antibunching in weakly interacting Bose and Fermi gases, respectively. Here we report the use of this probe to study correlations in a range from 60 to 180 nm in an atomic gas with strong interactions. Exploiting the large s -wave scattering length of ^{86}Sr ($a_{86} = 811 a_0$ [22], where a_0 is the Bohr radius), we access a regime where interactions dominate and the pair correlation function reflects universal few-body behavior. Our measurements show clear suppression of $g^{(2)}(R)$ in a strongly interacting gas of ^{86}Sr compared

to the relatively weakly interacting isotope ^{84}Sr ($a_{84} = 123 a_0$ [23]) and agree quantitatively with *ab initio* calculations using a finite-range two-body interaction.

ULRRMs [18,19] consist of one or more ground-state atoms bound to a Rydberg atom through electron-atom scattering. Measurement of spatial correlations with ULRRMs takes advantage of the nearly constant bond length associated with small vibrational amplitudes in ULRRM dimers in the ground vibrational state [20]. Therefore, the dimer photoassociation probability is proportional to the probability of finding an atom pair in the initial gas with separation close to the bond length $R_n \approx 1.8(n - \delta)^2 a_0$, where n is the Rydberg principal quantum number and δ is the quantum defect. By scanning the principal quantum number, $g^{(2)}(R)$ of the initial gas can be mapped. Earlier studies of ULRRM formation in weakly interacting ultracold gases of atomic strontium ^{84}Sr and ^{87}Sr revealed bunching and Pauli exclusion in the two-body [20] and three-body [21] correlation functions as a result of Bose and Fermi statistics, respectively.

In ^{86}Sr , a natural halo state exists on the ground-state potential, which causes an unusually large s -wave scattering length that is close to the internuclear separation for $n = 25$ ULRRM dimers. a_{86} is much larger than the van der Waals length of the ground-state potential ($\ell_{\text{vdW}} = (M_{86}C_6/\hbar^2)^{1/4} = 75 a_0$) [24], but is significantly less than the de Broglie wavelength $\lambda_{\text{dB}} = 3200 a_0$ for the sample at $T = 1.3 \mu\text{K}$, where M_{86} is the atomic mass and C_6 is the long-range van der Waals coefficient for the atom-atom potential [25]. This enables examination of a universal regime [26,27], where the correlation function is determined by a single parameter, a_{86} , neglecting three-body effects. Comparison with the more weakly interacting ^{84}Sr provides normalization and allows us to assess numerical calculations and the range of validity of approximations of the correlation functions [1] in strongly interacting Bose gases.

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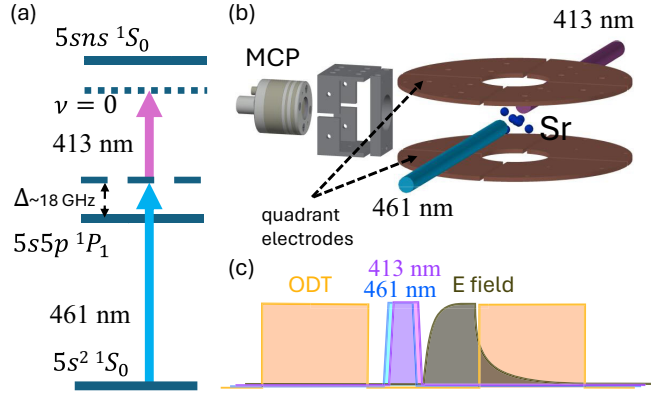


FIG. 1. ULRRM spectroscopy. (a) $5s^2 1S_0 \rightarrow 5sns 1S_0$ ULRRM photoexcitation scheme, (b) simplified diagram of the inside of the science chamber, and (c) ULRRM photoexcitation and detection pulse sequence used.

II. EXPERIMENTAL METHODS

The experimental setup is described in detail in previous work [20,21]. ^{84}Sr and ^{86}Sr atoms (both bosons with nuclear spin $I = 0$) are laser cooled via a two-staged magneto-optical trap that utilizes the $5s^2 1S_0 \rightarrow 5s5p 1P_1$ and then $5s^2 1S_0 \rightarrow 5s5p 3P_1$ transitions. After transfer to a crossed optical dipole trap (ODT), atoms are further cooled to a final temperature of 1.3 μK through free evaporation at a constant trap depth. The prepared samples typically contain 10^6 atoms at a peak density of $3 \times 10^{12} \text{ cm}^{-3}$.

ULRRMs are produced in both samples in the vibrational ground state using a two-photon transition ($5s^2 1S_0 \rightarrow 5s5p 1P_1 \rightarrow 5sns 1S_0$) driven by counterpropagating 461- and 413-nm photons with aligned polarizations [see Figs. 1(a) and 1(b)]. Both lasers are stabilized to a high-finesse cavity constructed with ultra low expansion glass, resulting in a total excitation bandwidth of less than 50 kHz. To limit off-resonant scattering, the 461-nm photons are 18 GHz blue-detuned from resonance with the intermediate state. The magnetic field is zeroed to less than 10 mG. The 413-nm photons are tuned to select ULRRMs with Rydberg principal quantum number $28 \leq n \leq 47$. Considering the quantum defects for $5sns 1S_0$ Rydberg states is $\delta = 3.269$ [28], this range of n corresponds to ULRRM sizes $58 \text{ nm} \lesssim R_n \lesssim 182 \text{ nm}$.

Rydberg molecules are produced using laser pulses of 20- μs duration. After each pulse, an electric field ramp is applied to induce field ionization (Fig. 1). Liberated electrons are guided to a microchannel plate (MCP) for detection. Excitation rates are limited to typically one Rydberg per laser pulse or fewer to avoid effects of Rydberg-Rydberg interactions. This experimental cycle is repeated 100 times on a single trapped sample at each two-photon excitation energy. At the end of these cycles, the atom number and temperature are measured by releasing atoms from the trap and monitoring cloud expansion via resonant absorption-imaging on the $5s^2 1S_0 \rightarrow 5s5p 1P_1$ transition. (Temperature and atom number vary by less than 10% during the 100 excitation pulses, and this small variation is neglected.) Then a new sample is trapped and the process is repeated for the next photoassociation energy. Typical measured spectra are shown in Fig. 2.

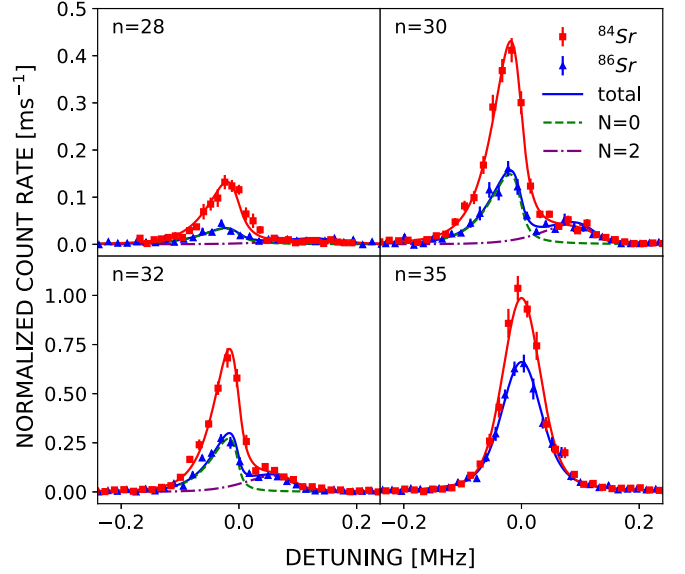


FIG. 2. Excitation spectra for $5sns 1S_0 v = 0$ ULRRM for ^{84}Sr and ^{86}Sr as a function of detuning from resonance for an atom pair initially at rest. The signals are normalized as described in the text. Note the different scales for $n = 28$ and 30 and $n = 32$ and 35 . Solid lines (^{84}Sr red, ^{86}Sr blue) show fits using a two-component convolution line shape described by Eq. (1) ($n = 28, 30$, and 32), and a simple Voigt profile ($n = 35$). Dashed lines show the contributions from $N = 0$ and $N = 2$ rotational states in ^{86}Sr . Zero detuning corresponds to the molecular binding energy of the $N = 0$ rotational state.

The normalization procedure used to account for drifts in the experimental parameters is described below. For calibration purposes, we also record atomic Rydberg excitation spectra in a similar fashion.

III. RESULTS AND DISCUSSION

A theoretical model of the photoexcitation line shape for singlet ULRRM dimers, including the effects of interactions in the atomic gas, was presented elsewhere [29,30]. This work showed that the excitation rate for a total two-photon laser detuning ω to a given vibrational level v is proportional to a sum over final ULRRM rotational states and a thermally weighted average over initial atom scattering states for temperature T and reduced mass μ , written as

$$f(\omega) \propto \frac{I_1 I_2}{\Delta^2} \beta_n \sqrt{\frac{2}{\pi}} \left(\frac{\hbar^2}{\mu k_B T} \right)^{3/2} \sum_{N, M_N} \int dk k^2 e^{-\hbar^2 k^2 / (2\mu k_B T)} \times \sum_{N', M'_N} |F_{v, N, M_N}(k, N', M'_N)|^2 L_{v, N}(k, \omega). \quad (1)$$

The total angular momentum and projection quantum numbers for initial and final states are labeled as N' and M'_N and N and M_N , respectively, and $\hbar k$ is the initial relative momentum of the colliding pair. $F_{v, N, M_N}(k, N', M'_N)$ is the Frank-Condon overlap between the initial and final states. Only even values of N are allowed for the indistinguishable bosons used here. The excitation laser intensities are I_1 and I_2 and the intermediate state detuning is Δ . β_n is the square of

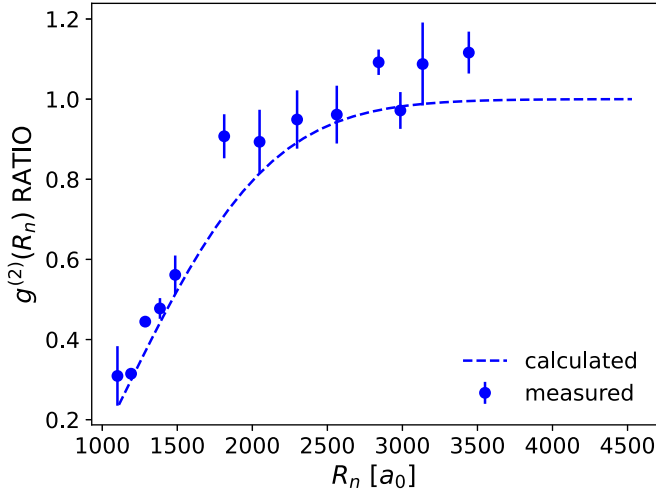


FIG. 3. The ratio of nonlocal correlation functions $g_{86}^{(2)}(R)/g_{84}^{(2)}(R)$ determined experimentally from the ratio of the normalized spectral integrals $\Gamma_{n,v=0}$ together with the results of theoretical calculations as described in the text.

the reduced two-photon electronic-transition matrix element, which varies with principal quantum number as n^{-3} . The spectrum is also convolved with the Lorentzian instrumental line shape $L_{v,N}(k, \omega)$ that takes into account energy conservation and Doppler-broadening, which is negligible in this study. Rotational states are resolved for $n \lesssim 33$. At larger n (and inter-nuclear separation), the rotational energy becomes small and a simple Voigt profile fits the dimer spectrum well. Figure 2 shows typical spectra and fits using Eq. (1), along with contributions from $N = 0$ and $N = 2$ rotational states.

The correlation function is extracted from the integral, $\Gamma_{n,v=0}$, of the experimental signals summed over all rotational levels. Accounting for factors that are required to normalize the signal, $\Gamma_{n,v=0}$ is proportional to the detection efficiency α , the integral over space of the square of the atom density $\rho(\mathbf{r})$, and the integral over frequency of the excitation rate $f(\omega)$:

$$\begin{aligned} \Gamma_{n,v=0} &= \alpha \int d^3\mathbf{r} \rho(\mathbf{r})^2 \int d\omega f(\omega) \\ &\propto \alpha \frac{I_1 I_2}{\Delta^2} N \rho_0 \beta_n O_n g^{(2)}(R_n). \end{aligned} \quad (2)$$

The integral over space and density yields a numerical factor times the product of the total atom number N and the peak density ρ_0 . Because of the localization of the ULRRM vibrational wave function at internuclear separation near R_n , the frequency integral of $f(\omega)$ is proportional to $g^{(2)}(R_n)$, the pair-correlation function for separation R_n , and an n -dependent factor O_n that can be accurately calculated from knowledge of the ULRRM vibrational wave functions (see Appendix B) [20].

To account for variations in experimental parameters, spectra (e.g., Fig. 2) are normalized by I_1 , I_2 , N , and ρ_0 for each trapped sample [31]. By taking the ratio between the normalized $\Gamma_{n,v=0}$ for ^{86}Sr and ^{84}Sr , common factors like α , O_n , and β_n are eliminated. This yields the ratio $g_{86}^{(2)}(R)/g_{84}^{(2)}(R)$ shown in Fig. 3. Figure 3 also shows the ratio calculated numerically from the expectation values of the two-particle, reduced

density matrix for ^{86}Sr and ^{84}Sr using a local-density approximation and partial-wave-scattering wave functions that include the s -wave-scattering phase shifts. (See Appendix A and Eq. 4.23 of Ref. [1].) The decrease in this ratio seen as the interparticle separation decreases is a result of strong interactions on $g_{86}^{(2)}(R)$. The ratios of the measured and calculated $g^{(2)}(R)$ curves agree reasonably well with no adjustable parameters. Systematic deviations are within the bounds expected from systematic uncertainties in the density and temperature measurements for the two isotopes. Three-body effects have been neglected in this analysis [26,32].

If one assumes that the nonlocal spatial correlation function for ^{84}Sr follows the form for a noninteracting Bose gas, $g_0^{(2)}(R) = 1 + e^{-2\pi(R/\lambda_{\text{dB}})^2}$, then $g_{86}^{(2)}(R)$ can be extracted from the data in Fig. 3 with no additional calculations or assumptions. However, as discussed below, there are indications that interactions have a small effect on $g^{(2)}(R)$ for ^{84}Sr at the lower end of the range of internuclear separations probed here.

In order to independently extract measured values of $g^{(2)}(R)$ for each isotope, the normalized $\Gamma_{n,v=0}$ signals are divided by the integral of the atomic Rydberg excitation spectrum for the same n , scaled to the same laser parameters and sample number and density. This removes dependence on α and β_n . We also scale the signal by the n dependence of $O_n \propto (n - \delta)^{6.7}$ calculated numerically from the overlap of the rovibrational ULRRM wave functions with initial scattering states, neglecting atom-atom interactions (Appendix B). This scaling agrees well with the dependence $O_n \propto (n - \delta)^{6.5 \pm 0.3}$ determined from the excitation rates observed for ULRRM dimers in a ten-state mixture of ^{87}Sr , which approximates a classical gas [20].

The resulting data match for both isotopes for $R \gtrsim \lambda_{\text{dB}}$, providing confirmation of the interisotope calibration factors applied in this analysis. A single scaling factor is then applied to set the average of the values of $g^{(2)}(R_n)$ for $n > 42$ for ^{84}Sr and ^{86}Sr to unity. This yields the measured $g^{(2)}(R_n)$ from ULRRM dimer spectroscopy for ^{84}Sr and ^{86}Sr shown in Fig. 4.

$g_{84}^{(2)}(R)$ for the weakly interacting isotope ^{84}Sr increases as the interparticle separation decreases, as expected for bosonic bunching. For comparison, Fig. 4 includes the correlation function for an ideal Bose gas, $g_0^{(2)}(R)$, which rises towards 2 for $R \ll \lambda_{\text{dB}}$. Figure 4 also shows numerical calculation of $g_{84}^{(2)}(R)$ from the expectation value of the two-particle, reduced density matrix (Appendix A), which reveals that even for the moderate values of the s -wave scattering length for ^{84}Sr [23] ($a_{84} = 124 a_0 \approx \lambda_{\text{dB}}/25$), spatial correlations are suppressed due to atom-atom interactions. Reference [1] provides an intuitive analytical approximation for the correlation function for an interacting gas that is valid for $a \ll \lambda_{\text{dB}}/\sqrt{2\pi}$:

$$\begin{aligned} g^{(2)}(R) &= g_0^{(2)}(R) \left(1 - \frac{a}{R}\right)^2 \\ &\quad + \{1 - [g_0^{(1)}(R)]^2\} \left[\left(1 + \frac{a}{R}\right)^2 - 1\right], \end{aligned} \quad (3)$$

where $g_0^{(1)}(R) = e^{-\pi(R/\lambda_{\text{dB}})^2}$ is the first-order correlation function for a noninteracting Bose gas, and $(1 - \frac{a}{R})^2$ is the square of the relative wave function in the limit of low scattering energy. For ^{84}Sr , Eq. (3) agrees well with the numerical

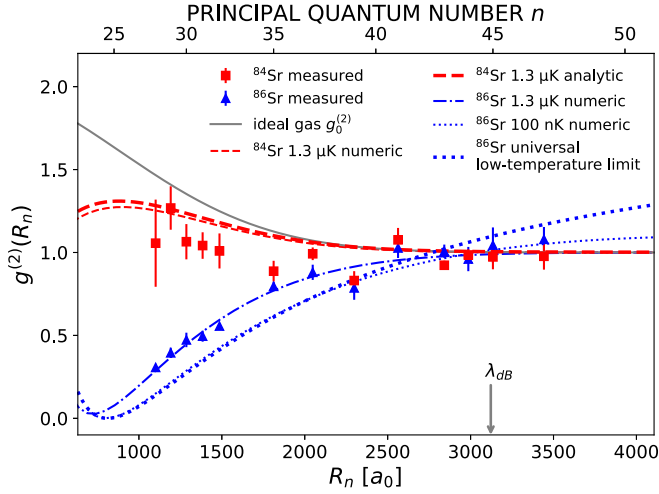


FIG. 4. Comparison of measured values of the nonlocal spatial correlation function $g^{(2)}(R)$ for ^{84}Sr and ^{86}Sr with theoretical calculations, including for an ideal gas, numerical calculations taking interactions into account using the two-body reduced density matrix (Appendix A), analytical expressions valid for weak interactions [Eq. (3)], and the universal low-temperature limit [Eq. (4)]. The arrow indicates the de Broglie wavelength for ^{86}Sr atoms at 1.3 μK .

calculation, and the second term adds a 5% correction at most. Experimental data are systematically below the ideal gas values for $R/\lambda_{\text{dB}} \lesssim 0.5$ and are reasonably consistent with predictions including interactions, although we note that the deviations from ideal gas behavior are on the order of uncertainties in the applied scaling for O_n .

In contrast to ^{84}Sr , $g_{86}^{(2)}(R)$ shows strong suppression as R decreases, reflecting the large scattering length $a_{86} = 811 a_0$ [22] of ^{86}Sr . Because the scattering length satisfies $\ell_{\text{vdW}} \ll a_{86} \ll \lambda_{\text{dB}}$, this behavior has an intuitive explanation. For $R \ll \lambda_{\text{dB}}$, spatial correlations are sensitive only to the s -wave-scattering partial wave, which for ^{86}Sr has a node at $R = a_{86} \approx 0.25\lambda_{\text{dB}}$, causing $g_{86}^{(2)}(R)$ to vanish there. Experimental values match the prediction of $g_{86}^{(2)}(R)$ based on numerical calculation of the reduced two-particle density matrix over the entire accessible range.

The analytical expression based on Eq. (3), however, yields a nonphysical result for ^{86}Sr , notably missing the strong suppression of $g^{(2)}(R)$ near the scattering length. At the present sample temperature and for ^{86}Sr , the assumption underlying the theory—that the scattering length is much smaller than the de Broglie wavelength—is not satisfied, and the second term in Eq. (3) is large.

In the zero-temperature limit, because of the increased λ_{dB} , Eq. (3) is valid over the range of experimentally measured internuclear separations, and it reduces to

$$g^{(2)}(R) \approx g_0^{(2)}(R) \left(1 - \frac{a}{R}\right)^2 \approx 2 \left(1 - \frac{a}{R}\right)^2, \quad (4)$$

which is proportional to the square of the magnitude of the scattering wave function. To investigate the significance of thermal effects on the form of $g_{86}^{(2)}(R)$, Fig. 4 shows the numerical prediction for $T = 100$ nK, which matches the zero-temperature limit.

This demonstrates that the present experimental measurements lie very close to the universal limit [Eq. (4)] in which the form of $g^{(2)}(R)$ is determined by a single parameter, the s -wave scattering length. Such behavior is only accessible for a gas with strong interactions such that the scattering length is large enough to dominate the behavior over a wide range of experimentally accessible internuclear separations.

IV. CONCLUSION

We have used photoassociation of ULRRMs to perform *in situ* measurements of the nonlocal two-body spatial correlation function $g^{(2)}(R)$ in ^{86}Sr , a Bose gas with strong interactions. This method probes length scales from 60 to 180 nm, where the large scattering length of ^{86}Sr results in a pronounced suppression of spatial correlations, and the correlation function approaches a universal form at sufficiently low temperatures. *Ab initio* calculations based on the two-body reduced density matrix match the data without adjustable parameters.

These measurements establish ULRRMs as a powerful experimental platform for exploring equilibrium many-body physics with spatial resolution that bridges the gap between conventional short-range probes and optical-scale imaging. The technique can be readily extended to other atomic species and interaction regimes, and to the study of nonequilibrium dynamics, opening new avenues for tests of many-body theory and exploration of exotic phases of matter.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [33].

APPENDIX A: NUMERICAL CALCULATIONS OF TWO-BODY CORRELATIONS

The pair-correlation functions are calculated for a gas of atoms at low density for which three-body and higher-order collisions are negligible. (At the peak density the probability to find two atoms within a distance equal to the scattering length ($\sim 811 a_0$) is about 3×10^{-3} and that for three atoms is 9×10^{-6} .) In this limit the pair correlation can be approximated [1] as

$$G^{(2)}(\vec{r}_1, \vec{r}_2) = \langle \vec{r}_1, \vec{r}_2 | \Omega^\dagger G_0^{(2)} \Omega | \vec{r}_1, \vec{r}_2 \rangle. \quad (A1)$$

For two-body scattering, the Møller operator Ω maps a state before the collision $|\vec{k}_1, \vec{k}_2\rangle$ onto an asymptotic state after the collision.

When the asymptotic state is assumed to have a constant scattering amplitude a , i.e.,

$$\langle \vec{r}_1, \vec{r}_2 | \Omega | \vec{k}_1, \vec{k}_2 \rangle = e^{i\vec{k}_{\text{CM}} \cdot \vec{R}_{\text{CM}}} \left(e^{i\vec{k} \cdot \vec{R}} - \frac{a}{R} e^{ikR} \right), \quad (A2)$$

the pair correlation of interacting bosons can be expressed analytically by Eq. (3) [1]. $\vec{R}_{\text{CM}} = (\vec{r}_1 + \vec{r}_2)/2$ and $\vec{R} = \vec{r}_1 - \vec{r}_2$ are the center of mass and relative coordinates, and $\vec{k}_{\text{CM}} = \vec{k}_1 + \vec{k}_2$ and $\vec{k} = (\vec{k}_1 - \vec{k}_2)/2$ are the total and relative wave vectors. This approximation is valid only for two-body scattering with small scattering length $a \ll \lambda_{dB}/\sqrt{2\pi}$.

For the numerical calculations presented here estimating the pair-correlation function, we replace the scattering wave function [Eq. (A2)] with the two-body wave function obtained numerically using the finite-range interaction potential [25]. (Note that the potential is for the $^1S + ^1S$ asymptote of Sr_2 , but the same potential is employed also for higher partial waves.) The calculated wave functions indicate that the momentum-dependent s -wave scattering length $a(k)$ stays within 10% of its zero-energy value $a(0)$ up to $T = k^2/(\mu k_B) = 2.4 \mu\text{K}$, even for ^{86}Sr with $a(0) = a_{86} \simeq 811$ a.u. Consequently, a node in the scattering wave functions around $R = a_{86}$ survives even after thermal averaging at $T = 1.3 \mu\text{K}$ and the pair correlation displays a clear suppression.

APPENDIX B: n -DEPENDENT SCALING FACTORS IN THE EXCITATION STRENGTH

For ultra-long-range Rydberg molecules, the interaction between the Rydberg electron and the ground-state atom is typically very weak. Therefore, the electronic wave function of a Rydberg molecule is essentially the same as that of a Rydberg atom. Therefore, the excitation strength can be factorized to the squared two-photon dipole transition matrix element of the electron, β_n , and an effective Franck-Condon factor, $\mathcal{F}$. The two-photon dipole transition scales as $\beta_n \propto (n - \delta)^{-3}$. The Franck-Condon factor between the molecular state $\langle \vec{R} | \nu, N, M_N \rangle = \mathcal{R}_{\nu,N}(R) Y_N^{M_N}(\hat{R})$ and the single initial scattering state $\langle \vec{R} | k, N', M'_N \rangle = j_{N'}(kR) Y_{N'}^{M'_N}(\hat{R})$ is given by

$$F_{\nu,N,M_N}(k, N', M'_N) = \delta_{N,N'} \delta_{M_N,M'_N} \int dR R^2 \mathcal{R}_{\nu,N}(R) j_{N'}(kR). \quad (\text{B1})$$

We note that for excitation of strontium singlet $5sns$ Rydberg states, the effects of recoil momentum [29] are small and neglected. The effective Franck-Condon factor $\mathcal{F}$ for the ground vibrational state ($\nu = 0$) becomes proportional to the thermal average of the Franck-Condon factor summed over rotational levels, N and M_N , i.e.,

$$\mathcal{F} \propto \sum_N (2N+1) \sqrt{\frac{2}{\pi}} \left(\frac{\hbar}{\mu k_B T} \right)^{3/2} \int dk k^2 e^{-\hbar^2 k^2 / (2\mu k_B T)} \times |F_{\nu=0,N,M_N}(k, N, M_N)|^2. \quad (\text{B2})$$

For bosons, the effective Franck Condon factor $\mathcal{F}_{\text{boson}}$ is obtained by summing over rotational states with even N . For ^{84}Sr and ^{86}Sr the spherical Bessel function $j_{N'}(kR)$ is phase-shifted

only for $N' = 0$ to account for the s -wave scattering length. The effective Franck-Condon factor $\mathcal{F}_{cl,n}$ is also evaluated for an ideal gas of distinguishable atoms and used to normalize the excitation strength. One key feature is that the ground vibrational state has a radial wave function well localized around $R = R_n \simeq 1.8(n - \delta)^2$ and nearly N -independent $\mathcal{R}_{\nu,N}(R) \simeq \mathcal{R}_\nu(R)$. At low temperature, $\lambda = 2\pi/k$, is much longer than the width of the vibrational wave function and the spherical Bessel function can be treated as nearly constant around $R = R_n$. Within this approximation the effective Franck-Condon factor can be factorized as

$$\mathcal{F} \propto \left[\sum_N (2N+1) \sqrt{\frac{2}{\pi}} \left(\frac{\hbar}{\mu k_B T} \right)^{3/2} \times \int dk k^2 e^{-\hbar^2 k^2 / (2\mu k_B T)} |j_{N'}(kR_n)|^2 \right] \times \left| \int dR R^2 \mathcal{R}_{\nu,N}(R) \right|^2. \quad (\text{B3})$$

The first bracket is the thermal average of the initial scattering state which is proportional to the pair-correlation function of the initial gas. The pair correlation of bosons is obtained by summing over the rotational states with even N and that of fermions is obtained by summing over the rotational states with odd N . Normalization is given by $g^{(2)}(R = R_n) \simeq \mathcal{F}_{\text{boson}}/O_n$, where

$$O_n \simeq \left| \int dR R^2 \mathcal{R}_{\nu,N}(R) \right|^2 \quad (\text{B4})$$

is an n -dependent factor that depends on the molecular wave function. Because the factor in brackets in Eq. (B3) equals unity if the sum extends over all N , O_n approximates the effective Franck-Condon factor $\mathcal{F}_{cl,n}$ for an ideal gas, $O_n \simeq \mathcal{F}_{cl,n}$ which is the n -dependent excitation strength. Numerical calculations confirm that O_n agrees with $\mathcal{F}_{cl,n}$ for low n or low temperature.

The factorization of the effective Franck-Condon factor is only strictly valid for relatively small values of n because the vibrational wave function for $n > 50$ becomes gradually delocalized and the n dependence of the factor O_n needs to be more carefully determined. Even when the vibrational wave function is delocalized, the extracted value can be interpreted as the average pair correlation over the width of the vibrational wave function $\mathcal{R}_{\nu=0}$ around $R \simeq R_n$. O_n can be calculated accurately from knowledge of the effective potential for ULRRMs and is found to vary as $O_n \propto (n - \delta)^{6.7}$ in the range of n (< 50) used here, while the exponent increases to $O_n \propto (n - \delta)^{7.0}$ for $n > 50$.

- [1] M. Naraschewski and R. J. Glauber, *Phys. Rev. A* **59**, 4595 (1999).
- [2] A. Browaeys and T. Lahaye, *Nat. Phys.* **16**, 132 (2020).
- [3] G. Semeghini, H. Levine, A. Keesling, S. Ebadi, T. T. Wang, D. Bluvstein, R. Verresen, H. Pichler, M. Kalinowski, R. Samajdar,

- A. Omran, S. Sachdev, A. Vishwanath, M. Greiner, V. Vuletić, and M. D. Lukin, *Science* **374**, 1242 (2021).
- [4] A. Mazurenko, C. S. Chiu, G. Ji, M. F. Parsons, M. Kanász-Nagy, R. Schmidt, F. Grusdt, E. Demler, D. Greif, and M. Greiner, *Nature (London)* **545**, 462 (2017).

- [5] R. A. Hart, P. M. Duarte, T.-L. Yang, X. Liu, T. Paiva, E. Khatami, R. T. Scalettar, N. Trivedi, D. A. Huse, and R. G. Hulet, *Nature (London)* **519**, 211 (2015).
- [6] B. Paredes, A. Widera, V. Murg, O. Mandel, S. Fölling, I. Cirac, G. V. Shlyapnikov, T. W. Hänsch, and I. Bloch, *Nature (London)* **429**, 277 (2004).
- [7] B. Laburthe Tolra, K. M. O'Hara, J. H. Huckans, W. D. Phillips, S. L. Rolston, and J. V. Porto, *Phys. Rev. Lett.* **92**, 190401 (2004).
- [8] T. Kinoshita, T. Wenger, and D. S. Weiss, *Phys. Rev. Lett.* **95**, 190406 (2005).
- [9] E. Haller, M. Rabie, M. J. Mark, J. G. Danzl, R. Hart, K. Lauber, G. Pupillo, and H.-C. Nägerl, *Phys. Rev. Lett.* **107**, 230404 (2011).
- [10] K. V. Kheruntsyan, D. M. Gangardt, P. D. Drummond, and G. V. Shlyapnikov, *Phys. Rev. Lett.* **91**, 040403 (2003).
- [11] E. A. Burt, R. W. Ghrist, C. J. Myatt, M. J. Holland, E. A. Cornell, and C. E. Wieman, *Phys. Rev. Lett.* **79**, 337 (1997).
- [12] M. Schellekens, R. Hoppeler, A. Perrin, J. V. Gomes, D. Boiron, A. Aspect, and C. I. Westbrook, *Science* **310**, 648 (2005).
- [13] K. M. Jones, E. Tiesinga, P. D. Lett, and P. S. Julienne, *Rev. Mod. Phys.* **78**, 483 (2006).
- [14] R. Yao, S. Chi, M. Wang, R. J. Fletcher, and M. Zwierlein, *Phys. Rev. Lett.* **134**, 183402 (2025).
- [15] T. de Jongh, J. Verstraten, M. Dixmierias, C. Daix, B. Peaudecerf, and T. Yefsah, *Phys. Rev. Lett.* **134**, 183403 (2025).
- [16] J. Xiang, E. Cruz-Colón, C. C. Chua, W. R. Milner, J. de Hond, J. F. Fricke, and W. Ketterle, *Phys. Rev. Lett.* **134**, 183401 (2025).
- [17] W. S. Bakr, J. I. Gillen, A. Peng, S. Fölling, and M. Greiner, *Nature (London)* **462**, 74 (2009).
- [18] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, *Phys. Rev. Lett.* **85**, 2458 (2000).
- [19] V. Bendkowsky, B. Butscher, J. Nipper, J. P. Shaffer, R. Löw, and T. Pfau, *Nature (London)* **458**, 1005 (2009).
- [20] J. D. Whalen, S. K. Kanungo, R. Ding, M. Wagner, R. Schmidt, H. R. Sadeghpour, S. Yoshida, J. Burgdörfer, F. B. Dunning, and T. C. Killian, *Phys. Rev. A* **100**, 011402(R) (2019).
- [21] S. K. Kanungo, Y. Lu, F. B. Dunning, S. Yoshida, J. Burgdörfer, and T. C. Killian, *Phys. Rev. A* **107**, 033322 (2023).
- [22] J. A. Aman, J. C. Hill, R. Ding, K. R. A. Hazzard, T. C. Killian, and W. Y. Kon, *Phys. Rev. A* **98**, 053441 (2018).
- [23] Y. N. Martinez de Escobar, P. G. Mickelson, P. Pellegrini, S. B. Nagel, A. Traverso, M. Yan, R. Côté, and T. C. Killian, *Phys. Rev. A* **78**, 062708 (2008).
- [24] C. Chin, R. Grimm, P. Julienne, and E. Tiesinga, *Rev. Mod. Phys.* **82**, 1225 (2010).
- [25] A. Stein, H. Knöckel, and E. Tiemann, *Eur. Phys. J. D* **57**, 171 (2010).
- [26] F. Werner and Y. Castin, *Phys. Rev. A* **86**, 053633 (2012).
- [27] P. Makotyn, C. E. Klauss, D. L. Goldberger, E. A. Cornell, and D. S. Jin, *Nat. Phys.* **10**, 116 (2014).
- [28] R. A. Brienza, Y. Lu, C. Wang, S. K. Kanungo, T. C. Killian, F. B. Dunning, J. Burgdörfer, and S. Yoshida, *Phys. Rev. A* **108**, 022815 (2023).
- [29] Y. Lu, J. D. Whalen, S. K. Kanungo, T. C. Killian, F. B. Dunning, S. Yoshida, and J. Burgdörfer, *Phys. Rev. A* **106**, 022809 (2022).
- [30] C. Wang, Y. Lu, S. K. Kanungo, F. B. Dunning, T. C. Killian, and S. Yoshida, *Phys. Rev. A* **110**, 032803 (2024).
- [31] C. Wang, Y. Lu, A. Li, S. K. Kanungo, T. C. Killian, F. B. Dunning, and S. Yoshida, *Phys. Rev. A* **111**, 052811 (2025).
- [32] R. J. Wild, P. Makotyn, J. M. Pino, E. A. Cornell, and D. S. Jin, *Phys. Rev. Lett.* **108**, 145305 (2012).
- [33] C. Wang, Non-local spatial correlations in an interacting atomic gas, version 1.0, Harvard Dataverse (2025), <https://doi.org/10.7910/DVN/7K67EO>.