

Supplemental material

Special case: Additive interaction

In the special case of additive interactions where the Hilbert space factorizes fully into orthonormal single-particle subspaces, further simplifications occur. If we let $\{|b\rangle\}$ and $\{|c\rangle\}$ be orthonormal bases spanning subsystems B and C , respectively, then the many-body projector P_k can be written as

$$P_k = \binom{N}{k} Q_B^{\otimes k} \otimes Q_C^{\otimes(N-k)}, \quad (\text{S1})$$

where the single-particle projectors onto B and C are

$$Q_B = \sum_b |b\rangle\langle b|, \quad Q_C = \sum_c |c\rangle\langle c|. \quad (\text{S2})$$

This immediately yields a factorized expression for $S_k(t)$ in terms of single-particle overlaps, greatly simplifying the evaluation of $S(t)$ to

$$S(t) = \sum_{k=0}^N \binom{N}{k} [s_{k,B}(t)]^k [s_{k,C}(t)]^{N-k} = [s(t)]^N, \quad (\text{S3})$$

where in the last step the binomial theorem was used to recover the known expression [1, 2]

$$S(t) = \left| \sum_{\alpha} |\langle \psi_0 | \psi_{\alpha} \rangle|^2 e^{-it(\varepsilon_{\alpha} - \varepsilon_0)} \right|^N, \quad (\text{S4})$$

with $\varepsilon_{\alpha}, |\psi_{\alpha}\rangle$ the eigenenergy and eigenstate of the interacting Hamiltonian of a single boson with the impurity.

In Fig. 1, we compare the truncated k -subspace expansion from Eq. S3 (up to $k = 2$) with the full solution, following Eq. S4, for an isotropic and additive interaction in a Rydberg s -state. The full absorption spectrum (Fig. 1a) exhibits a smooth broadening across each resonance, reflecting the cumulative contribution of all many-body states. In contrast, the truncated spectrum (Fig. 1b) shows a breakdown of this continuity: the attractive-polaron feature no longer smoothly connects to the repulsive side, and the Gaussian-like envelope near unitarity disappears entirely. Instead, only discrete peaks associated with dimer and trimer molaron states remain visible. This highlights that higher-order cluster contributions—clearly present in the full calculation—are absent when truncating the expansion at low k , and thus

a breakdown of this description at strong couplings. However, away from this region, the truncated description excellently captures the full absorption spectrum including the line-shifts and broadening due to continuum scattering and spectral weight of the different absorption features.

An intuitive derivation of $S(t)$

Our goal in this section is to derive an approximate expression for $S(t)$ based on a hierarchical treatment of the possible M -body bound states. The approach presented here is less rigorous but provides a more intuitive understanding of the underlying structure. To do so, we begin with the additive case where we can expand $S(t)$ into two-body states of the interacting and non-interacting system, $|\gamma\rangle$ and $|0\rangle$ respectively, yielding

$$\begin{aligned} S(t) &= \left[\sum_{\gamma} |\langle 0 | \gamma \rangle|^2 \exp\{i(\omega_0 - \epsilon_{\gamma})t\} \right]^N \\ &= \left[\sum_{\gamma} s_{\gamma} \right]^N \end{aligned} \quad (\text{S5})$$

Next we separate the two-particle states into bound states $|\beta\rangle$ and continuum states $|\alpha\rangle$, and neglect all correlations between bound states, meaning $(\sum_{\gamma} s_{\gamma})^M = (\sum_{\beta} s_{\beta}^M) + (\sum_{\alpha} s_{\alpha})^M$. This leads to

$$\begin{aligned} S(t) &= \sum_{M=1}^N \binom{N}{M} \left(\left[\sum_{\beta} s_{\beta}^M \right] \left[\sum_{\gamma} s_{\gamma} \right]^{N-M} \right. \\ &\quad \left. + \left[\sum_{\alpha} s_{\alpha} \right]^M \left[\sum_{\gamma} s_{\gamma} \right]^{N-M} \right). \end{aligned} \quad (\text{S6})$$

Since the contribution of bound states is much smaller than that of continuum states, we can set $s_{\gamma} \approx s_{\alpha}$, in the limit of $M \ll N$, which results in

$$S(t) = \sum_{M=0}^N \binom{N}{M} \left[\sum_{\beta} s_{\beta}^M \right] \left[\sum_{\alpha} s_{\alpha} \right]^{N-M} \quad (\text{S7})$$

Here the first term gives a molaron, with the bound states $|\beta\rangle$ populated by M many bosons which gets dressed with continuum distributions $|\alpha\rangle$ of $(N - M)$ bath particles.

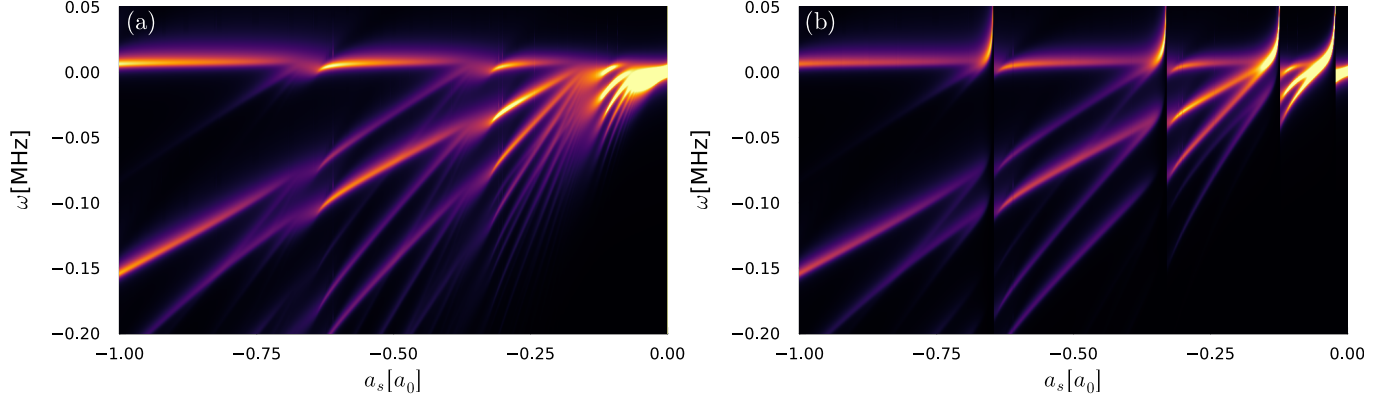


Figure 1: Absorption spectra for a Rydberg $|50s\rangle$ state as a function of the electron–atom scattering length a_s . (a) Full calculation of the absorption $A(\omega)$ using the Loschmidt echo in Eq. S4. (b) Effect of truncating the cluster expansion in Eq. S3 at $k = 2$, showing how limiting the sum alters the spectral features, especially close to unitarity.

However, for a non-additive many-body system, the occupation-dependent basis changes dynamically. Specifically, the number of bound-state particles, M , directly modifies the full basis. Therefore, for a non-additive potential, we must compute the basis states independently for each term in the sum over M ,

$$S(t) = \sum_{M=0}^N \binom{N}{M} \sum_{\beta} (s_{\beta_M}) \left[\sum_{\alpha(\beta_M)} s_{\alpha(\beta_M)} \right]^{N-M}. \quad (\text{S8})$$

Here we also phenomenologically include that the continuum states depend on the underlying molecular state. Assuming that all bound and continuum states are mutually orthogonal simplifies the symmetrization of the total bosonic wavefunction and allows the above factorization; however, this approximation underlies the entire derivation and is essential for the validity of the final expression.

Details for non-additive Hamiltonians

For calculating the polaron contribution with $k = 0$, the continuum scattering states are obtained using $\hat{H}_C^{k,\alpha,\gamma}$ with the spherically symmetric electronic state

$$|\phi_{\gamma}(\vec{R}_i)|^2 = \left| \frac{u_{n1}(R_i)}{R_i} Y_{10}(0,0) \right|^2. \quad (\text{S9})$$

This reflects the fact that the electronic orbital can freely localize along whichever direction an incoming bath par-

ticle approaches. The bound states that arise in this potential correspond to the $k = 1$ contribution. Once a molecule forms, however, the electron localizes into an $\ell = 1, m = 0$ state, thereby imposing an anisotropic potential for subsequent scattering of the remaining bath particles—analogueous to the effect of an external magnetic field.

To solve for the $k = 2$ case, we start by solving Eq. 2 for two bath particles, which will give rise to two non-zero Born-Oppenheimer potential energy surfaces. These surfaces lack a simple analytical form and must be computed numerically to accurately determine the bare trimer energies. While such calculations are possible, our present goal is to gain qualitative insight into non-additive effects and these are not sensitive to the precise values of the binding energies. Therefore, for simplicity, we approximate the trimer interaction (following the approach of [3]) by assuming that the radial and angular dependencies of the BO potentials factorize. For the radial part, we assume that the two bath particles do not influence each other directly, so the radial dependence is identical to that of the dimer case. The angular dependence is then obtained by setting $R_1 = R_2 = R$ and integrating out the electronic degrees of freedom as a function of Θ , the relative angle between the two ground state atoms. Under these assumptions, the two non-zero BO surfaces take the

form

$$V_{\pm}^{\text{BO}}(R, \Theta) = 2\pi a_s \left| \frac{u_{nl}(R)}{R} \right|^2 \cdot \frac{3}{4\pi} \left(1 \pm \frac{1}{\sqrt{3}} \cos(\Theta) \right). \quad (\text{S10})$$

For each bound-state $\langle \vec{R}_1 \vec{R}_2 | \Psi_{\alpha, \gamma}^{(k)} \rangle_B = \Psi_B^{\alpha, \gamma}(\vec{R}_1, \vec{R}_2)$ of this Hamiltonian we then have to solve for the continuum dressing. For this we calculate the spatial expectation value for each molecular state which, due to the simple single-well structure of each of the two potential surfaces (as a function of Θ), is always in the well center. Here the corresponding state of the electron is a pure $l = 1, m = 0$ state. Thus, each trimer state is attached to a electronic localization in the same state as the dimer. This is an outcome of all the simplifying approximations employed in the trimer calculation, and is neither a universal feature or necessary for the approach.

Using this we can then solve for the single particle continuum state $\psi_C^{k, \alpha, \gamma, y}(\vec{R})$. As a next step we need to evaluate the Loschmidt echo for the triatomic partition, $s_2(t)$. Here one needs to be careful, as the bound and continuum states are not mutually orthogonal $\langle \Psi_B^{\alpha, \gamma}(\vec{R}_1, \vec{R}_2) | \psi_C^{2, \alpha, \gamma, a}(\vec{R}_1) \psi_C^{2, \alpha, \gamma, b}(\vec{R}_2) \rangle \neq 0$. This leads to a total $S(t = 0) > 1$, indicating that the space we constructed is overcomplete. To fix this, we orthogonalize the continuum states with respect to the bound states. This first requires that we construct a set of symmetric two-boson continuum states $\Psi_C^{2, \alpha, \gamma, y=(a,b)}(\vec{R}_1, \vec{R}_2)$. The symmetric two-body bosonic continuum state, where the particles occupy the single particle states $\psi_C^{2, \alpha, \gamma, a}$ and $\psi_C^{2, \alpha, \gamma, b}$, is

$$\Psi_C^{2, \alpha, \gamma, y=(a,b)}(\vec{R}_1, \vec{R}_2) = \frac{1}{\sqrt{2(1 + \delta_{ab})}} \left[\psi_C^{2, \alpha, \gamma, a}(\vec{R}_1) \psi_C^{2, \alpha, \gamma, b}(\vec{R}_2) + \psi_C^{2, \alpha, \gamma, a}(\vec{R}_2) \psi_C^{2, \alpha, \gamma, b}(\vec{R}_1) \right]. \quad (\text{S11})$$

In a Gram-Schmidt process we then create the set of orthogonal states, $\{\Psi\} = \{\Psi_1^B, \Psi_2^B, \dots, \tilde{\Psi}_1^C, \tilde{\Psi}_2^C, \dots\}$, where all $\tilde{\Psi}_i^C$ are linear combinations of Ψ^B and Ψ^C as a result

of the orthogonalization. The n^{th} state is given as

$$\begin{aligned} |\tilde{\Psi}_n^C\rangle &= |\Psi_n^C\rangle - \sum_j^{N_B} \frac{\langle \Psi_j^B | \Psi_n^C \rangle}{\langle \Psi_j^B | \Psi_j^B \rangle} |\Psi_j^B\rangle \\ &\quad - \sum_i^n \frac{\langle \tilde{\Psi}_i^C | \Psi_n^C \rangle}{\langle \tilde{\Psi}_i^C | \tilde{\Psi}_i^C \rangle} |\tilde{\Psi}_i^C\rangle, \\ &= |\Psi_n^C\rangle - \sum_j^{N_B} \frac{\langle \Psi_j^B | \Psi_n^C \rangle}{\langle \Psi_j^B | \Psi_j^B \rangle} |\Psi_j^B\rangle, \end{aligned} \quad (\text{S12})$$

with the normalization

$$d_i = \sqrt{\langle \tilde{\Psi}_i^C | \tilde{\Psi}_i^C \rangle} = \sqrt{1 - \sum_j |\langle \Psi_j^B | \tilde{\Psi}_i^C \rangle|^2} \quad (\text{S13})$$

such that $\langle \Psi_j^B(\vec{R}_1, \vec{R}_2) | \tilde{\Psi}_i^C(\vec{R}_1, \vec{R}_2) \rangle = 0$.

As these orthogonal states are now linear combinations of eigenstates of the Hamiltonian, we calculate their energies following

$$H |\tilde{\Psi}_n^C\rangle = \sum_i E_i \langle \Psi_i^{C,B} | \tilde{\Psi}_n^C \rangle |\Psi_i^{C/B}\rangle. \quad (\text{S14})$$

With this the Loschmidt echo takes the form

$$\begin{aligned} s_2(t) &= \sum_{\alpha} \sum_{\gamma} \left[\langle \Psi_0 | \Psi_{2, \alpha, \gamma}^B \rangle e^{-i(E_{k, \alpha, \gamma}^B - N\epsilon_0)t} \langle \Psi_{2, \alpha, \gamma}^B | \Psi_0 \rangle \right. \\ &\quad \cdot \left(\sum_{y, y'} \langle \Psi_0 | \Psi_{2, \alpha, \gamma, y}^C \rangle e^{-i(E_y - E_0)t} \right. \\ &\quad \left. \left. \langle \Psi_{2, \alpha, \gamma, y}^C | \tilde{\Psi}_{2, \alpha, \gamma, y'}^C \rangle \langle \Psi_0 | \tilde{\Psi}_{2, \alpha, \gamma, y'}^C \rangle \right) \right]^{\frac{N-k}{k}} \end{aligned} \quad (\text{S15})$$

Together, these steps provide a consistent definition of $s_2(t)$ that incorporates both bound and continuum contributions while avoiding overcounting due to the non-orthogonality of the original states. **Extension to the $k = 3$ sector would follow similar lines, but with significant added complexity stemming from the higher dimensionality of the tetramer problem.**

Magnetic fields

The Rydberg-boson two-body system in a magnetic field B is described by the Hamiltonian

$$\hat{H} = \hat{H}_{\text{Ryd}}(\vec{r}) - \left[\frac{\nabla_{\vec{R}}^2}{2\mu} - 2\pi a_s \delta^3(\vec{r} - \vec{R}) - \vec{B} \cdot \hat{\vec{L}} \right], \quad (\text{S16})$$

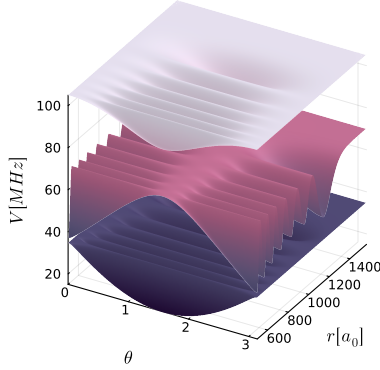


Figure 2: Potential energy surfaces of a Rydberg p -state and a ground state atom, split by a magnetic field.

with $\vec{R}$ the internuclear distance and $\vec{r}$ the electronic coordinate. In the standard Born-Oppenheimer approach, we first solve the electronic problem by treating the interaction with a ground-state atom as a perturbation on top of a hydrogenic Rydberg state characterized by quantum numbers n , l , and m :

$$\begin{aligned} & \langle nlm | B\vec{L}_z - 2\pi a_s \delta(\vec{r} - \vec{R}) | n'l'm' \rangle \\ & \approx \langle nlm | B\vec{L}_z - 2\pi a_s \delta(\vec{r} - \vec{R}) | nlm' \rangle \\ & = Bm\delta_{m,m'} + 2\pi a_s \left| \frac{u_{nl}(R)}{R} \right|^2 Y_{lm}^*(\Omega) Y_{lm'}(\Omega). \end{aligned} \quad (\text{S17})$$

The magnetic field term Bm lifts the degeneracy of the magnetic sublevels, as illustrated in Fig. 2 for a p -state Rydberg atom. In this regime, each electronic state is energetically well separated from the others, allowing for selective excitation without significant m -level mixing. In what follows, we focus on the $l = 1$, $m = 0$ state.

In order to find the eigenfunctions of the nuclear Hamiltonian we expand the vibrational wave function $\psi(R, \Omega)$ into spherical harmonics as well,

$$\psi(R, \Omega) = \sum_{L,M} Y_{LM}(\Omega) \frac{\chi_{LM}(R)}{R}. \quad (\text{S18})$$

By projecting out the LM -dependence of the Hamilto-

nian we arrive at a system of coupled radial equations

$$\begin{aligned} \langle LM | \hat{H} | \psi \rangle &= \left(-\frac{1}{2\mu} \frac{d^2}{dR^2} + \frac{L(L+1)}{2\mu R^2} \right) \chi_{LM}(R) \\ &+ \sum_{L',M'} 2\pi a_s \left| \frac{u_{nl}(R)}{R} \right|^2 \chi_{L'M'} \langle LM | Y_{lm}^* Y_{lm} | L'M' \rangle. \end{aligned} \quad (\text{S19})$$

Since the interaction term is block-diagonal in M , and we assume a spherically symmetric initial state with $M = 0$, we restrict our analysis to this symmetry sector and drop the explicit M -dependence in what follows. Including the parity of the spherical harmonics, the angular momenta overlap in the interaction of Eq. S19 is only non-zero if L, L' are both even or both odd, which further reduces the basis dimensions.

The dynamics of this full many-body problem is described by the Loschmidt echo

$$S(t) = \left(\sum_{\gamma} |\langle \psi_0 | \psi_{\gamma} \rangle|^2 \exp\{-it(\epsilon_0 - \epsilon_{\gamma})\} \right)^N \quad (\text{S20})$$

where $|\psi_0\rangle$, ϵ_0 is lowest non-interacting eigenstate and its energy, and $|\psi_{\gamma}\rangle$, ϵ_{γ} is a interacting state and its energy of the interacting two-body system given by Eq. S19. In order to solve this we need to calculate the overlap between each of the eigenstates $|\gamma\rangle$ and the spherically symmetric non-interacting state

$$\psi_0(R, \Omega) = Y_{00}(\Omega) \frac{\chi_0(R)}{R}. \quad (\text{S21})$$

The radial solution is given by $\chi_0(R) = \sin[R\pi/R_{\text{box}}]$ with an appropriate normalization. Due to the orthogonality of the spherical harmonics the overlap integral simplifies to

$$\begin{aligned} \langle \psi_0 | \psi_{\alpha} \rangle &= \int dR d\Omega Y_{00}^*(\Omega) \chi_0(R) \sum_{\ell} Y_{\ell 0}(\Omega) \chi_{\alpha, \ell}(R) \\ &= \sum_{\ell} \delta_{\ell, 0} \int dr \chi_0(R) \chi_{\alpha, \ell}(R) \end{aligned} \quad (\text{S22})$$

the $\ell = 0$ component.

For this calculation we include all scattering channels corresponding to even L up to $L_{\text{max}} = 40$ and expand the radial wave function into a basis composed of 3000 radial B-splines distributed over a knot grid spanning $[300, 10^5]a_0$ with a_0 being the Bohr-radius. A diagonalization of the coupled channel equations (S19) then yields the interacting scattering states $|\psi_{\alpha}\rangle$, from which we include around 1000 eigenstates in the summation in Eq. S20.

High densities

As the density is increased beyond $a_{\text{Ryd}} \approx \rho^{1/3}$ and bosons accumulate inside the potential, the details of individual states vanish and melt into a continuum of molaron states with a Gaussian envelope. In Fig. 3 the devolution of the absorption spectrum with increasing density for a Rydberg impurity in a magnetic field are shown. With increasing density, higher-order clusters (trimers and beyond) acquire increasing spectral weight. At the same time, the density of molecular states increases, leading to a reduced energy separation between individual features. Once this spacing becomes comparable to the intrinsic linewidth, the discrete peaks begin to overlap and merge into broader spectral features. In this regime, the molecular structure appears progressively “washed out,” forming a smooth, approximately Gaussian envelope centered near the many-body shifted energy. This behavior is consistent with previous studies [2, 4, 5] of long-range Rydberg and ionic impurities, where increasing density enhances the occupation of higher-order clusters and redistributes spectral weight away from few-body states.

Remarkably, the universal feature arising at high density – characterized by zero quasiparticle weight – is not specific to any particular quantum state but arises purely from pressure broadening in the unitarity regime of finite-range interactions in impurity systems. This universality implies that the energy of the spectral feature depends only on the bath density and boson species, independent of the Rydberg electron’s quantum numbers. In the

mean-field picture, this energy can be expressed as:

$$E_{\text{MF}} = 2\pi a_s \rho \int dr |u_{nl}(r) Y_{l,m}(\vartheta, \varphi)|^2 = 2\pi a_s \rho. \quad (\text{S23})$$

Since the Rydberg electron’s wave function is normalized, the high-density spectral feature appears identical across all Rydberg electronic states, highlighting its fundamental nature. This aligns perfectly with the findings for Rydberg s-state impurities [1, 5, 6].

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- [1] R. Schmidt, J. D. Whalen, R. Ding, F. Camargo, G. Woehl, S. Yoshida, J. Burgdörfer, F. B. Dunning, E. Demler, H. R. Sadeghpour, and T. C. Killian, *Physical Review A* **97**, 022707 (2018).
 - [2] A. A. T. Durst and M. T. Eiles, *Physical Review Research* **6**, L042009 (2024).
 - [3] C. Fey, J. Yang, S. T. Rittenhouse, F. Munkes, M. Baluktsian, P. Schmelcher, H. Sadeghpour, and J. P. Shaffer, *Physical Review Letters* **122**, 103001 (2019).
 - [4] E. R. Christensen, A. Camacho-Guardian, and G. M. Bruun, *Physical Review Letters* **126**, 243001 (2021).
 - [5] R. Schmidt, H. Sadeghpour, and E. Demler, *Physical Review Letters* **116**, 105302 (2016).
 - [6] F. Camargo, R. Schmidt, J. Whalen, R. Ding, G. Woehl, S. Yoshida, J. Burgdörfer, F. Dunning, H. Sadeghpour, E. Demler, and T. Killian, *Physical Review Letters* **120**, 083401 (2018).

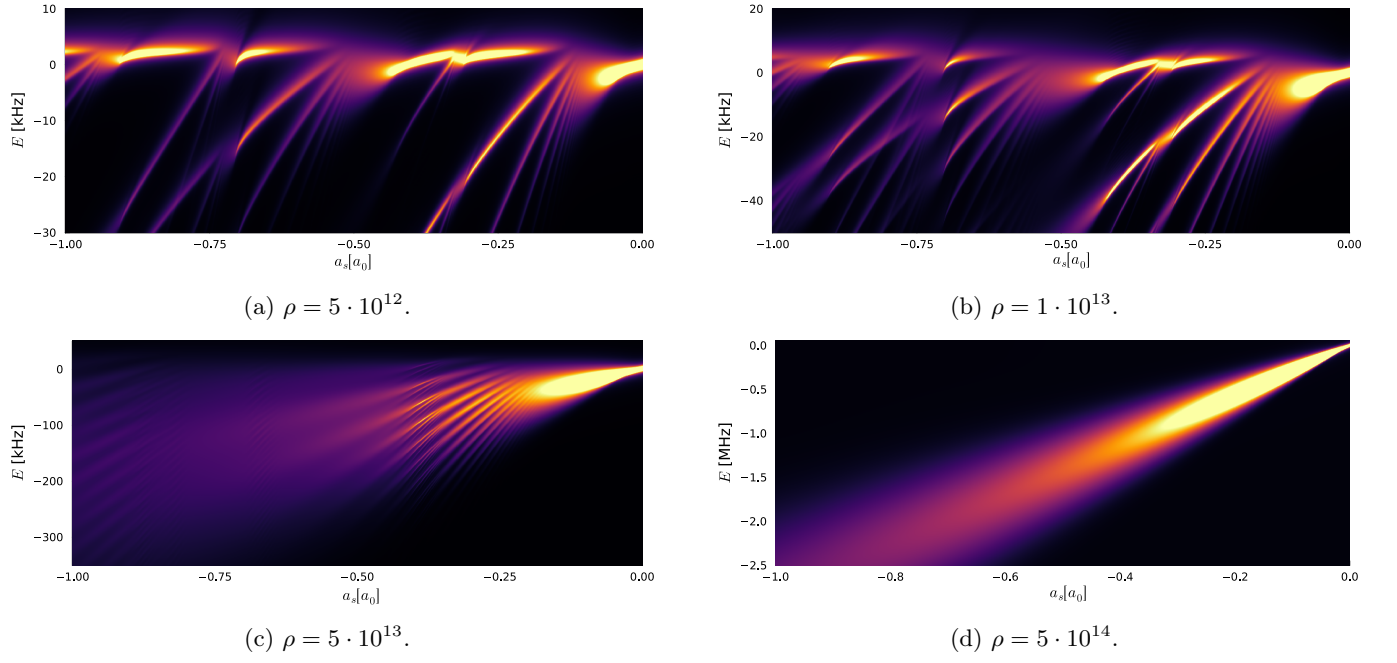


Figure 3: Comparison of the absorption spectra of a Rydberg $|n = 50, l = 1, m = 0\rangle$ state immersed into a BEC in the order of increasing gas density ((a) – (d)) in a strong magnetic field.