LETTERS

Repulsively bound atom pairs in an optical lattice

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Throughout physics, stable composite objects are usually formed by way of attractive forces, which allow the constituents to lower their energy by binding together. Repulsive forces separate particles in free space. However, in a structured environment such as a periodic potential and in the absence of dissipation, stable composite objects can exist even for repulsive interactions. Here we report the observation of such an exotic bound state, which comprises a pair of ultracold rubidium atoms in an optical lattice. Consistent with our theoretical analysis, these repulsively bound pairs exhibit long lifetimes, even under conditions when they collide with one another. Signatures of the pairs are also recognized in the characteristic momentum distribution and through spectroscopic measurements. There is no analogue in traditional condensed matter systems of such repulsively bound pairs, owing to the presence of strong decay channels. Our results exemplify the strong correspondence between the optical lattice physics of ultracold bosonic atoms and the Bose–Hubbard model[1,2]—a link that is vital for future applications of these systems to the study of strongly correlated condensed matter and to quantum information.

Cold atoms loaded into a three-dimensional (3D) optical lattice provide a realization of a quantum lattice gas[1,2]. An optical lattice can be generated by pairs of counterpropagating laser beams, where the resulting standing wave intensity pattern forms a periodic array of microtraps for the cold atoms, with period a given by half the wavelength of the light, $\lambda/2$. The periodicity of the potential gives rise to a band structure for the atom dynamics with Bloch bands separated by bandgaps, which can be controlled by the laser parameters and beam configuration. The dynamics of ultracold atoms loaded into the lowest band of a sufficiently deep optical lattice is well described by the Bose–Hubbard model with hamiltonian[1,3]:

$$\hat{H} = -J\sum_{\langle i,j\rangle}\hat{b}_i^{\dagger}\hat{b}_j + \frac{U}{2}\sum_i \hat{n}_i(\hat{n}_i - 1) + \sum_i \varepsilon_i \hat{n}_i \qquad (1)$$

Here $\hat{b}_i$ ($\hat{b}_i^{\dagger}$) are destruction (creation) operators for the bosonic atoms at site i, and $\hat{n}_i = \hat{b}_i^{\dagger}\hat{b}_i$ is the corresponding number operator. $J/\hbar$ denotes the nearest-neighbour tunnelling rate, U the on-site collisional energy shift, and ε_i the background potential. The high degree of control available over the parameters in this system—for example, changing the relative values of U and J by varying the lattice depth, V_0—has led to seminal experiments on strongly correlated gases in optical lattices. These experiments include the study of the superfluid–Mott insulator transition[4], the realization of one-dimensional (1D) quantum liquids with atomic gases[5,6] (see also refs 7 and 8), and the investigation of disordered systems[9]. 3D optical lattices have also opened new avenues in cold collision physics and chemistry[10–13].

A striking prediction of the Bose–Hubbard hamiltonian (equation (1)) is the existence of stable repulsively bound atom pairs. These are most intuitively understood for strong repulsive interaction $|U| \gg J$, $U > 0$, where an example of such a pair is a state of two atoms occupying a single site, $|2_i\rangle \equiv (\hat{b}_i^{\dagger 2}|\mathrm{vac}\rangle)/\sqrt{2}$, where $|\mathrm{vac}\rangle$ is the vacuum state. This state has a potential energy offset U with respect to states where the atoms are separated (Fig. 1a). The pair is unable to decay by converting the potential energy into kinetic energy, as the Bloch band allows a maximum kinetic energy for two atoms given by $8J$, twice its width. The pair can move around the lattice, with both atoms tunnelling to a neighbouring site (Fig. 1b), but the atoms cannot move independently. The stability of repulsively bound pairs is intimately connected with the absence of dissipation, in contrast to solid state lattices, for example, where interactions with phonons typically lead to rapid relaxation.

We obtain experimental evidence for repulsively bound pairs with a sample of ultracold ^{87}Rb atoms in a cubic 3D optical lattice with lattice period $a = 415.22$ nm. The key tool used to prepare and observe the pairs is their adiabatic conversion into chemically

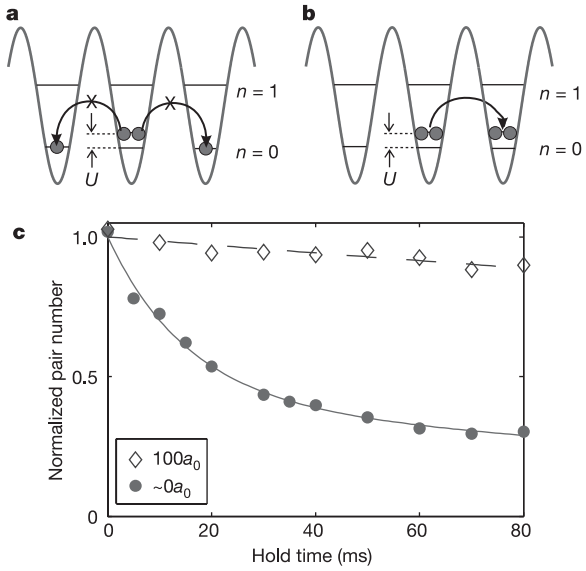

Figure 1 | Atom pairs in an optical lattice. **a**, Repulsive interaction (scattering length $a > 0$) between two atoms sharing a lattice site in the lowest band ($n = 0$) gives rise to an interaction energy U. Breaking up of the pair is suppressed owing to the lattice band structure and energy conservation. **b**, The pair is a composite object that can tunnel through the lattice. **c**, Long lifetime of repulsively bound atom pairs that are held in a 3D optical lattice. The potential depth is $(10 \pm 0.5)E_r$ in one direction and $(35 \pm 1.5)E_r$ in the perpendicular directions. Shown is the remaining fraction of pairs for a scattering length of $100a_0$ (open diamonds; a_0 is the Bohr radius) and a scattering length of about $(0 \pm 10)a_0$ (filled circles) as a function of the hold time. The lines are fitted curves of an exponential (dashed line) and the sum of two exponentials (solid line).

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bound dimers using a magnetic-field sweep across a Feshbach resonance[13–20] at 1,007.40 G. The initial state is prepared from a pure sample of Rb_2 Feshbach molecules in the vibrational ground state of the lattice where each lattice site is occupied by not more than a single molecule (see Methods). Sweeping across the Feshbach resonance, we adiabatically dissociate the dimers and obtain a lattice correspondingly filled with 2×10^4 atom pairs, at an effective filling factor of typically 0.3. Away from the Feshbach resonance, the effective interaction between the atoms is repulsive with scattering length $a_s = +100a_0$ (where a_0 is the Bohr radius).

In order to demonstrate the stability of repulsively bound pairs, we lower the lattice potential in one direction from its initial depth of $V_0 = 35E_r$ (corresponding to $J/\hbar \approx 2\pi \times 0.7$ Hz and $U/J \approx 3{,}700$, where $E_r = 2\pi^2\hbar^2/m\lambda^2$ and m is the mass of the atoms) in 1 ms to a depth of $V_0 = 10E_r$. This increases dramatically the tunnelling rates along this direction to $J/\hbar \approx 2\pi \times 63$ Hz ($U/J \approx 30$), potentially allowing the atom pairs to quickly separate. After a variable hold time we determine the number of remaining pairs. This is done by adiabatically raising the lattice to its full initial depth of $V_0 = 35E_r$, and converting doubly occupied sites to Feshbach molecules with near unit efficiency[13]. A purification pulse[13] then removes all remaining atoms due to dissociated pairs. Afterwards the molecules are again converted back into atoms, and can then be detected by conventional absorption imaging.

The results of these lifetime measurements are shown in Fig. 1c. For repulsive interaction ($a_s = 100a_0$), the atom pair sample exhibits the remarkably long lifetime of 700 ms (exponential fit). This lifetime is mainly limited by inelastic scattering of lattice photons[13], and greatly exceeds the calculated time for an atom to tunnel from one site to the next, $2\pi\hbar/(4J) \approx 4$ ms. In contrast, if we turn off the on-site interaction by tuning the scattering length near zero, we observe a much faster decay in the number of doubly occupied sites owing to the rapid diffusion of unbound atoms through the lattice (Fig. 1c). This observation clearly demonstrates that the stability of the pairs is induced by the on-site interaction U.

We can more deeply understand these repulsively bound pairs through an exact solution of the two-particle Lippmann–Schwinger scattering equation based on the Bose–Hubbard model. We write the two-atom wavefunction as $\Psi(\mathbf{x}, \mathbf{y})$, where the positions of the two particles are denoted $\mathbf{x} = \Sigma_i x_i \mathbf{e}_i$ and $\mathbf{y} = \Sigma_i y_i \mathbf{e}_i$, with $\mathbf{e}_i$ being the primitive lattice vectors, and x_i, y_i integer numbers. Introducing centre of mass, $\mathbf{R} = (\mathbf{x} + \mathbf{y})/2$, and relative coordinates, $\mathbf{r} = \mathbf{x} - \mathbf{y}$, we can solve the Schrödinger equation with the ansatz $\Psi(\mathbf{x}, \mathbf{y}) = \exp(i\mathbf{KR})\psi_K(\mathbf{r})$, where $\mathbf{K}$ is the quasi-momentum of the centre of mass motion and $\psi_K(\mathbf{r})$ is the pair wavefunction. We derive two types of solutions (for details see Methods), each of which are eigenstates of $\mathbf{K}$. These states, as illustrated in Fig. 2a, correspond to (1) unbound scattering solutions (shaded area in Fig. 2a), where the two particles move on the lattice, and scatter from each other according to the interaction U, and (2) repulsively bound pairs for which $\psi_K(\mathbf{r})$ is square integrable. In one and two dimensions, states of repulsively bound pairs always exist for non-zero U, while in three dimensions they exist above a critical value $U_{crit} \approx 0.5J$.

In this Letter, we focus primarily on the 1D situation, which in the experiment corresponds to a low depth of the lattice along one direction, whilst the lattice in the perpendicular directions remains very deep ($35E_r$). Here the energy of the bound pairs is $E(K) = 2J\left[\sqrt{4(\cos\frac{Ka}{2})^2 + (U/2J)^2} + 2\right]$. This is plotted in Fig. 2a as the Bloch band of a stable composite object above the continuum of two-particle scattering states. In the limit of strong interaction, $U \gg J$, this reduces to $E(K) \approx 4J + U + (4J^2/U)(1 + \cos Ka)$, which shows that the bound pairs indeed have binding energy of $\sim U$ and hop through the lattice with an effective tunnelling rate $J^2/(\hbar U)$.

Figure 2b shows the pair wavefunctions $\psi_K(r)$ for repulsively bound pairs ($a_s = 100a_0$) in one dimension with $K = 0$, for $U/J = 30$ ($V_0 \approx 10E_r$) and $U/J = 3$ ($V_0 \approx 3E_r$). For large U/J, bound pairs essentially consist of two atoms occupying the same site, whereas for small U/J, the pair is delocalized over several lattice sites. The corresponding quasi-momentum distribution can be found from the Fourier transform $\tilde{\psi}_0(k)$ of the pair wavefunction (Fig. 2c), where k is the relative quasi-momentum. Because $K = 0$, $|\tilde{\psi}_0(k)|^2$ is also equal to the single-particle quasi-momentum distribution. When the two particles are localized on the same site, the quasi-momentum distribution is essentially flat. However, for lower U/J the wavefunction is characteristically peaked at the edges of the Brillouin zone. This occurs because the energy of the repulsively bound state is above that of the continuum, and thus the contribution to the corresponding wavepacket of single-particle quasi-momentum states with higher energy is favoured. In contrast, if we had $U < 0$, the pair would be attractively bound, and would have energy lower than that in the continuum. Thus contributions from the low-energy quasi-momentum states would be favoured, leading to a single peak in the centre of the Brillouin zone. In both cases, the amplitude of the peaks grows with increasing width $4J$ of the Bloch band. In general, the stable bound pairs will not be prepared in a fixed quasi-momentum state K in an experiment, but rather in a superposition of different momentum states. For non-zero K, the peaks in the single-particle quasi-momentum distribution are translated by K, but their strength is also reduced. As a consequence, for typical symmetric distributions of K, the peak at the edge of the Brillouin zone remains present, but is less strong than in the optimal case of vanishing K. We have verified this using many-body numerical simulations, which were performed using time-dependent density-matrix renormalization group methods[21–23].

We have experimentally investigated the quasi-momentum distribution of the pairs in various regimes by mapping it onto a spatial distribution, which we measured using standard absorption imaging. For this, we first adiabatically lower the lattice depth in the X

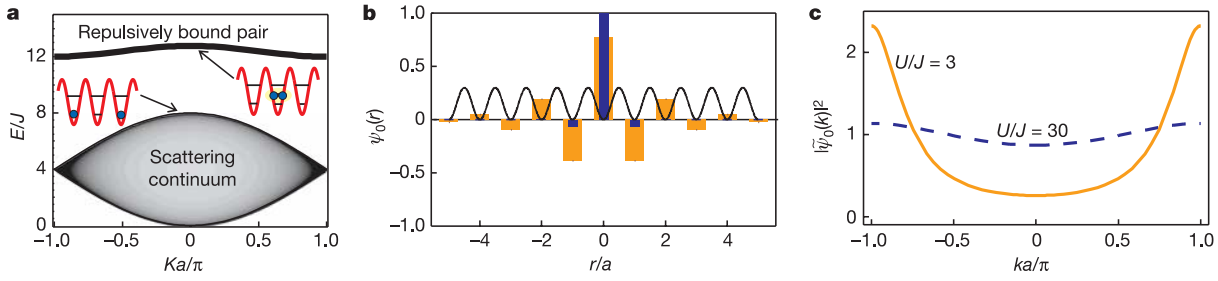

Figure 2 | Atom pair states in one dimension. **a**, Spectrum of energy E of the 1D hamiltonian for $U/J = 8$ ($V_0 \approx 6E_r$) as a function of centre of mass quasi-momentum K. The Bloch band for repulsively bound pairs is located above the continuum of unbound states. The grey level for the shading of the continuum is proportional to the density of states. **b**, The pair wavefunction $\psi_0(r)$, showing the amplitude at each site with $U/J = 30$ ($V_0 \approx 10E_r$, blue bars) and $U/J = 3$ ($V_0 \approx 3E_r$, orange bars). **c**, The square modulus of the corresponding momentum space wavefunctions $|\tilde{\psi}_0(k)|^2$, which are equivalent to the single-particle momentum distributions, as $K = 0$. Note the characteristic peaks at the edge of the Brillouin zone.

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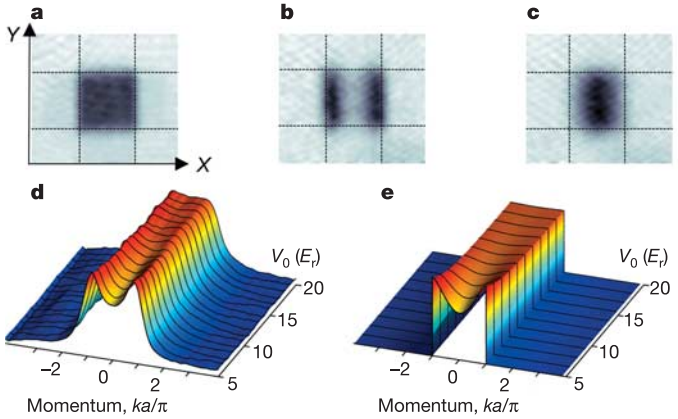

Figure 3 | Quasi-momentum distribution of atoms in the lattice. **a–c**, Absorption images of the atomic distribution after release from the 3D lattice and a subsequent 15-ms time of flight. The horizontal and vertical black lines enclose the first Brillouin zone. **a**, Distribution when lattice sites are occupied by single atoms; **b**, distribution for repulsively bound atom pairs (see text for details); **c**, same as **b** but pairs are attractively bound. **d**, **e**, The quasi-momentum distribution for pairs in the X direction as a function of lattice depth V_0, after integration over the Y direction. **d**, Experiment; **e**, numerical calculation. See Methods for a definition of E_r.

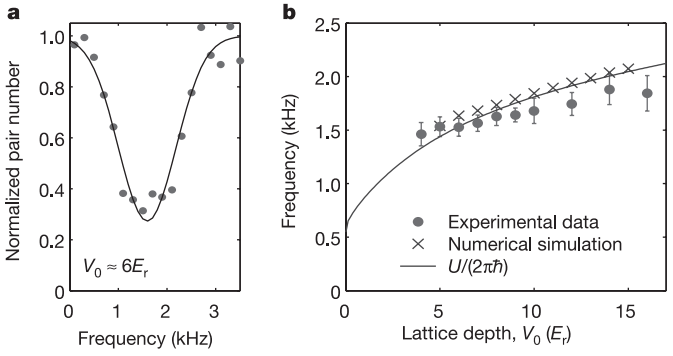

Figure 4 | Modulation spectroscopy of repulsively bound pairs. **a**, Typical resonance dip showing the remaining number of atom pairs as a function of the modulation frequency, for $V_0 \approx 6E_r$. The solid line is a gaussian fit, a choice that was justified by numerical calculations. **b**, Plot showing the measured resonance frequencies (filled circles) as a function of the lattice depth. They show good agreement with numerical simulations (crosses) and also coincide with the on-site collisional energy shift U (line). Experimental error bars correspond to the 95% confidence interval for the gaussian fit parameters of the resonance dips.

direction (Fig. 3a) at a rate of $1.3E_r$ (ms^{-1}) to a pre-chosen height while the lattice depth in the other two directions is kept high ($35E_r$). This will prepare repulsively bound pairs at the chosen lattice depth. We then turn off the lattice rapidly enough for the pair wavefunction not to change, but slowly with respect to the bandgap, so that single-particle quasi-momenta are mapped to real momenta[24,25]. We have typically employed linear ramps with rates of $0.2E_r\,\mu s^{-1}$. The resulting momentum distribution is converted to a spatial distribution after ~15 ms time of flight.

Figure 3a–c shows typical measured quasi-momentum distributions that were obtained after adiabatically lowering the lattice depth in the X direction to the lowest values, below $3E_r$. If only empty sites and sites with single atoms are present in the lattice, then the first Brillouin zone is homogeneously filled[24] (Fig. 3a). For repulsively bound pairs, the momentum distribution is, in general, peaked at the edges of the first Brillouin zone (Fig. 3b), whereas for attractively bound pairs, it is peaked in the centre of the first Brillouin zone (Fig. 3c). In order to change the interaction between the atoms from repulsive to attractive, we change the scattering length, making use of the Feshbach resonance[26] at 1,007.40 G. Figure 3d and e shows the dependence on lattice depth V_0 of the single-particle quasi-momentum distribution for repulsively bound pairs from experiment and numerical simulation, respectively. As expected, the peak structure is more pronounced for lower values of V_0, and diminishes for larger V_0. This characteristic is a clear signature of the pair wavefunction for repulsively bound pairs.

We also performed spectroscopic measurements, determining the binding energy from pair dissociation produced by modulating the depth of the lattice at a chosen frequency. On resonance, the modulation allows pairs to release their binding energy. Figure 4a shows the number of remaining pairs as a function of the modulation frequency. This was repeated for a variety of lattice depths V_0 in one direction while keeping the lattice in the other two directions at $35E_r$. The behaviour of the binding energy as a function of the lattice depth provides an additional key signature of repulsively bound pairs. As shown in Fig. 4b, the resonance positions are in good agreement with numerical simulations and essentially coincide with the interaction energy, U.

It is important to note that for sufficiently large U/J, repulsively bound pairs are stable under collisions with each other. This is particularly evident in the limit $U \gg J$ where, by energy arguments, the elastic scattering between pairs is the only open channel. This means that even a relatively dense quantum lattice gas of these objects can be long-lived. When the lattice height is lowered so that U/J becomes sufficiently small, it is possible for a certain fraction of the pairs to dissociate by collision with other pairs. In our experiments, we observe the onset of this behaviour for lattice depths lower than $6E_r$, that is, $U/J \approx 9$. The dynamics of the collisions and details of the decay depend crucially on lattice depth and the local density of pairs across the lattice. Further details of these processes will be discussed elsewhere.

In conclusion, we have demonstrated the formation of a novel composite object in an optical lattice: a stable bound state that arises from the lattice band structure and repulsion between the constituents. Although no direct analogue to repulsively bound atomic pairs is known to exist, the formation of a metastable state is reminiscent of trapping light in photonic bandgap materials[27], or extended lifetimes of excited atoms in cavity quantum electrodynamics[28]. In both cases, decay is suppressed by restriction of the accessible light field modes. Stable repulsively bound objects should be viewed as a general phenomenon, and their existence will be ubiquitous in cold atom lattice physics. They also give rise to new potential composites with fermions[29] or Bose–Fermi mixtures[30], and can be formed in an analogous manner with more than two particles. The stability of repulsively bound objects could thus be the basis of a wealth of new quantum many-body states or phases. In particular, the next experimental step in investigating repulsively bound atomic pairs is the possible realization of a condensate of pairs, together with the means to characterise long-range order in this system.

METHODS

Preparation of pure molecular sample. We use a set-up which was described in detail in ref. 13, starting with a Bose–Einstein condensate of 6×10^5 ^{87}Rb atoms in an Ioffe-type magnetic trap with trap frequencies $\omega_{x,y,z} = 2\pi \times (7, 19, 20)$ Hz). Within 100 ms the Bose–Einstein condensate is adiabatically loaded into the cubic 3D optical lattice which is $35E_r$ deep. After turning off the magnetic trap, we flip the spins of our atoms from their initial state $|F = 1, m_F = -1\rangle$ to $|F = 1, m_F = +1\rangle$ by suddenly reversing the bias magnetic field of a few gauss. This spin state features a 210-mG-wide Feshbach resonance at 1,007.40 G (ref. 26). By adiabatically ramping over this resonance we convert pairs of atoms in multiply occupied lattice sites into Rb_2 Feshbach molecules. Fast inelastic collisions of molecules within lattice sites and a subsequent combined radio-frequency and optical purification pulse remove all chemically unbound atoms, thus creating a pure molecular sample of about 2×10^4 molecules.

Exact solution for single pair bound state. Within the Bose–Hubbard model (equation (1)), the Schrödinger equation describing two particles in a

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homogenous optical lattice takes the form

$$\left[-J\left(\tilde{\Delta}_{\mathbf{x}}^{0}+\tilde{\Delta}_{\mathbf{y}}^{0}\right)+U\delta_{\mathbf{x},\mathbf{y}}\right]\Psi(\mathbf{x},\mathbf{y})=E\Psi(\mathbf{x},\mathbf{y}) \quad (2)$$

where the vectors $\mathbf{x}$ and $\mathbf{y}$ describe the positions of the two particles as defined in the main text. The operator $\tilde{\Delta}_{\mathbf{x}}^{\mathbf{K}}\Psi(\mathbf{x})=\sum_{i=1}^{d}\cos(\mathbf{K}\mathbf{e}_i/2)[\Psi(\mathbf{x}+\mathbf{e}_i)+\Psi(\mathbf{x}-\mathbf{e}_i)-2\Psi(\mathbf{x})]$ denotes the discrete lattice laplacian with d the dimensionality in the cubic lattice, and $\delta_{\mathbf{x},\mathbf{y}}$ is a Kronecker delta. Writing the wavefunction in relative and centre of mass coordinates $\Psi(\mathbf{x},\mathbf{y})=\exp(i\mathbf{K}\mathbf{R})\psi_K(\mathbf{r})$, the Schrödinger equation (2) then reduces to a single-particle problem in the relative coordinate

$$\left[-2J\tilde{\Delta}_{\mathbf{r}}^{\mathbf{K}}+E_{\mathbf{K}}+U\delta_{\mathbf{r},0}\right]\psi_K(\mathbf{r})=E\psi_K(\mathbf{r}) \quad (3)$$

with $E_{\mathbf{K}}=4J\Sigma_i[1-\cos(\mathbf{K}\mathbf{e}_i/2)]$ being the kinetic energy of the centre of mass motion.

The short range character of the interaction potential allows for a resummation of the perturbation expansion generated by the corresponding Lippmann–Schwinger equation. We obtain the scattering states

$$\psi^{(+)}(\mathbf{r})=\exp(i\mathbf{k}\mathbf{r})-8\pi J f_E(\mathbf{K})G_{\mathbf{K}}(E,\mathbf{r}) \quad (4)$$

with scattering amplitude

$$f_E(\mathbf{K})=-\frac{1}{4\pi}\frac{U/(2J)}{1-G_{\mathbf{K}}(E,0)U} \quad (5)$$

where the total energy $E=\varepsilon_{\mathbf{k},\mathbf{K}}+E_{\mathbf{K}}$, and $\varepsilon_{\mathbf{k},\mathbf{K}}=4J\Sigma_i\cos(\mathbf{K}\mathbf{e}_i/2)[1-\cos(\mathbf{k}\mathbf{e}_i)]$. Furthermore, $G_{\mathbf{K}}(E,\mathbf{r})$ denotes the Green's function of the non-interacting problem, which in Fourier space takes the form $\tilde{G}_{\mathbf{K}}(E,\mathbf{k})=1/(E-\varepsilon_{\mathbf{k},\mathbf{K}}+i\eta)$.

The energy spectrum for these states in one dimension is shown as a function of K by the shaded region in Fig. 2a. In addition, the pole in the scattering amplitude indicates the presence of an additional bound state. The energy E_{bs} of the bound state is determined by $G_{\mathbf{K}}(E_{\mathrm{bs}},0)U=1$ and the bound state wavefunction takes the form $\psi^{\mathrm{bs}}(\mathbf{r})=cG_{\mathbf{K}}(E_{\mathrm{bs}},\mathbf{r})$, with c being a normalization factor.

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