Controlling Correlated Tunneling and Superexchange Interactions with ac-Driven Optical Lattices

Yu-Ao Chen,* Sylvain Nascimbène, Monika Aidelsburger, Marcos Atala, Stefan Trotzky, and Immanuel Bloch

Fakultät für Physik, Ludwig-Maximilians-Universität, Schellingstrasse 4, 80799 München, Germany and Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, 85748 Garching, Germany

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The dynamical control of tunneling processes of single particles plays a major role in science ranging from Shapiro steps in Josephson junctions to the control of chemical reactions via light in molecules. Here we show how such control can be extended to the regime of correlated tunneling of strongly interacting particles. Through a periodic modulation of a biased tunnel contact, we have been able to coherently control single-particle and correlated two-particle hopping processes. We have furthermore been able to extend this control to superexchange spin interactions in the presence of a magnetic-field gradient. Such photon-assisted superexchange processes constitute a novel approach to realize arbitrary XXZ spin models in ultracold quantum gases, where transverse and Ising-type spin couplings can be fully controlled in magnitude and sign.

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The control of quantum tunneling of particles through a barrier using an oscillatory driving field lies at the heart of the interpretation of the so-called Shapiro steps observed in the I-V characteristics of a biased Josephson junction under an applied radio-frequency field [1,2]. Since then, other examples and applications of photon-assisted tunneling have emerged in several fields, such as the control of chemical reactions with laser pulses [3] or the observation of dynamic localization and absolute negative conductance in semiconductor superlattices [4]. Recently, the tunnel dynamics of single atoms in periodically modulated optical lattices was investigated, showing, in particular, the possibility to revert the sign of the tunnel coupling for strong driving amplitudes [5]. Periodically shaken optical lattices were also used as a spectroscopic tool for measuring the excitation spectrum of a superfluid Bose gas [6] or extracting nearest-neighbor spin correlations in a fermionic Mott insulator [7].

In this Letter, we demonstrate how the periodic modulation of an optical lattice can be used to selectively drive correlated tunneling processes. Hereby we focus on resonantly driven cotunneling and superexchange where in the absence of the driving both are inhibited by means of a potential energy offset or a magnetic-field gradient, respectively. The ability to resonantly drive superexchange processes eventually leads to a novel proposal to implement arbitrarily tunable XXZ spin models with ultracold atoms in optical lattices, where the anisotropy of the effective spin coupling can be adjusted by the strength of a magnetic-field gradient and the amplitude of the periodic driving alone. Existing proposals achieve the same only by changing the ratio of the interspecies and intraspecies interaction energies [8] which requires the existence of either a suitable Feshbach resonance or the use of near-detuned spin-dependent lattices accompanied by large heating rates [9].

The system under consideration is a lattice of isolated double-well potentials created by an optical superlattice [10,11]. A 3D optical lattice is formed by three mutually orthogonal retroreflected laser beams at the respective wavelengths $\lambda_{\text{sl}} = 1534$ nm ("long lattice"), $\lambda_{\text{s}} = 844$ nm, and $\lambda_{\text{l}} = 767$ nm. An additional standing wave with $\lambda_{\text{st}} \approx \lambda_{\text{s}}/2 \approx 767$ nm ("short lattice") is superimposed to the long lattice to obtain a superlattice of the form $V(x) = V_{s}\sin^2(k_{l}x) + V_{xs}\sin^2(2k_{l}x + \pi/2 - \phi)$, where $k_{l} = 2\pi/\lambda_{\text{st}}$ [see Fig. 1(a)]. The relative phase $\phi$ and the lattice depths $V_{\text{sl}}$ and $V_{\text{ss}}$ could be independently controlled in real time by dynamically adjusting the short-lattice wavelength $\lambda_{\text{ss}}$ and the laser intensities [11].

The vibrational level splitting being much larger than the other relevant energy scales, the system can be described by a two-site Hubbard-like Hamiltonian,

$$
\begin{align*}
H &= V_{s} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} + V_{xs} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} \cos \phi + V_{st} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} \sin \phi \\
&\quad - J_{\text{xx}} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} + J_{\text{zz}} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} \\
&\quad + J_{\text{xy}} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} \cos \phi + J_{\text{yx}} \sum_{\langle i,j \rangle} c_{i}^\dagger c_{j} \sin \phi,
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where $\langle i,j \rangle$ represents nearest-neighbor sites, $V_{s}$ is the on-site lattice potential, $J_{\text{xx}}$ and $J_{\text{zz}}$ are the nearest-neighbor hopping amplitudes along the $x$ and $z$ directions, respectively, $J_{\text{xy}}$ and $J_{\text{yx}}$ are the exchange interactions between the $x$- and $y$-oriented spins, and $V_{xs}$ is the amplitude of the superexchange.

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where \( J \) is the tunnel coupling, \( U \) is the on-site interaction energy, and \( \Delta \) is the potential tilt between the two sites. The last term represents a spin-dependent bias \( G \) provided by an additional magnetic-field gradient along the \( x \) direction. The operator \( \hat{a}_{R(L),\sigma} \) annihilates a particle in state \(|\sigma\rangle\) in the right (left) well, \( \hat{a}_{R(L),\sigma}^\dagger \hat{a}_{R(L),\sigma} \) is the corresponding number operator, and \( \hat{n}_{R(L)} = \sum_\sigma |a_{R(L),\sigma}|^2 \) is the population.

In addition to the static double-well potential, a time-periodic modulation was applied through a variation of the long-lattice depth \( V_{L}(t) = V_{0}^L + \delta V \cos(\omega t) \) [see Fig. 1(a)]. This modulation introduces an additional coupling term \( \hat{K} = -\sum_{\sigma = 1,0} \left[ g_{L}^\sigma \hat{a}_{L,\sigma}^\dagger \hat{a}_{R,\sigma} + g_{R}^\sigma \hat{a}_{R,\sigma}^\dagger \hat{a}_{L,\sigma} \cos(\omega t) \right] \) to the Hamiltonian (1), where the coupling constant \( g_{L}^\sigma \) is calculated using the Wannier functions \( w_{L,R}(x) \) localized in the left and right well, respectively [12]. For simplicity we neglect diagonal terms such as contributions proportional to \( \alpha_{L}^\dagger \alpha_{R} \). Each application of the operator \( \hat{K} \) changes the energy of the system by an amount \( \pm \hbar \omega \), which allows for the driving of tunneling processes which are forbidden by energy conservation in the bare Hamiltonian (1). It is convenient to picture this situation using the Floquet formalism [13] by dressing the atoms with effective “photons” of energy \( \hbar \omega \). Therefore, we denote the states of the driven Hamiltonian \( \hat{H} + \hat{K} \) by \( |L, R; \nu\rangle \), where \( L \) and \( R \) represent the left- and right-well populations and \( \nu \) is the effective photon or Floquet number.

Our experiments began by loading a Bose-Einstein condensate of about 10^5 87Rb atoms in the \(|F=1, m_F=-1\rangle\) Zeeman state into the 3D optical lattice formed by the long and transverse lattices alone. The final lattice depths were chosen so that the atomic sample was in the Mott insulating regime [14] with a central core of two atoms per well and an outer shell of singly occupied sites. We then transferred all atoms to the \(|F=1, m_F=0\rangle \equiv |0\rangle\) Zeeman state by a radio-frequency Landau-Zener adiabatic passage. Using microwave-dressed spin-changing collisions [15] we converted atom pairs in individual lattice sites into pairs with opposite magnetic moment, labeled as \(|\uparrow\rangle \equiv |F=1, m_F=-1\rangle\) and \(|\downarrow\rangle \equiv |F=1, m_F=1\rangle\). Single atoms resided in the state \(|0\rangle\).

In a first set of experiments, we loaded both single atoms and atom pairs to the “left” sides of asymmetric double wells by ramping up the short lattice with a phase of \( \phi = 0.18(1) \) rad. Direct tunneling to the right wells was inhibited by the tilt of the double wells. We then applied a modulation of the long-lattice depth with amplitude \( \delta V \) and frequency \( \omega \) for a time \( T \) to induce particle transfer. At the end of the modulation, we measured the populations \( n_{R_F} \) in the right wells by transferring them to a higher Bloch band and applying a subsequent band mapping technique [10,11,16]. To separate the different Zeeman states we used a magnetic-field gradient during time-of-flight imaging.

As shown by the black circles in Fig. 2, we observe a resonant transfer of single atoms (state \(|0\rangle\)) when the modulation frequency is equal to the energy difference between the two lowest eigenenergies of the Hamiltonian (1), i.e., \( \hbar \omega = \sqrt{\Delta^2 + 4F^2} = \Delta' \). In the Floquet picture, this is understood as a resonant coupling of the two states \(|1, 0; \nu\rangle\) and \(|0, 1; \nu - 1\rangle\) associated with the absorption of one photon of energy \( \hbar \omega = \Delta' \) from the driving field. The coherence of this process driven on resonance is illustrated in the inset of Fig. 2, where we plot \( n_{R_F} \) as a function of the modulation time \( T \). The single-particle scenario becomes modified if a second atom is present in the double well (state \(|\uparrow\rangle\), blue circles in Fig. 2). The initial state \(|\uparrow, 0; \nu\rangle\) is lifted by the interaction energy \( U \) compared to the single-particle state. From there, resonant transfer of single particles towards the entangled triplet

![FIG. 2 (color). Fraction of atoms transferred to the right well \( n_{R_F} \) as a function of the driving frequency \( \omega/2\pi \) with fixed modulation time \( T = 2.5 \) ms and driving amplitude \( \delta V = 8.2(3)E_F^{1/2} \) [24]. The lattice parameters given in the figure correspond to a tilt \( \Delta/h = 4.3(2) \) kHz. Single atoms (black circles) are resonantly transferred for \( \omega/2\pi = 5.2(1) \) kHz (gray vertical line), in agreement with \( \Delta' = h \times 5.0(2) \) kHz, where the value \( J/h = 1.30(5) \) kHz was obtained from independently measured single-particle tunnel oscillations in undriven symmetric double wells. In the case of atom pairs (blue circles), this resonance is shifted to \( \omega/2\pi = 3.9(1) \) kHz due to interactions. In both cases, the resonance around \( \omega/2\pi = 7.5 \) kHz can be attributed to a two-photon transfer to the third Bloch band. Finally, the resonance for atom pairs at \( \omega/2\pi = 10.8(2) \) kHz is \( 2\Delta'/h \) corresponds to the directly driven cotunneling of the pair. Inset: Time evolution of \( n_{R_F}(T) \) for the single-particle resonance measured with \( \delta V = 16.4(5)E_F^{1/2} \), together with a fit using a damped sine wave. The Rabi frequency provides the coupling \( K/h = 153(10) \) Hz, in reasonable agreement with the value of 166 (10) Hz obtained from a single-particle band structure calculation.](210405-2)
state $|\uparrow,\uparrow;\nu-1\rangle + |\downarrow,\downarrow;\nu-1\rangle)/\sqrt{2}$ occurs for a frequency $\hbar\omega < \Delta$ since in the final state the particles are spatially separated and practically do not interact.

In addition to the single-particle resonance, we observe a maximum in the transfer for atom pairs at $\omega/2\pi \approx 2\Delta / \hbar$, which we interpret as the coupling to the state $|0,\uparrow;\nu-1\rangle$. This coupling corresponds to the directly driven cotunneling of the atom pair [11,17] and has recently been explained as “fractional-photon assisted tunneling” [18]. We have verified this interpretation by performing spin-changing collisions on the final state, which are only effective if both particles are located in the same well. Further evidence comes from the fact that the resonance position is found to be insensitive to changes of $U$ induced by changing the transverse-lattice depths.

Having demonstrated resonantly driven tunneling and cotunneling with cold atoms inside a tilted double well, we now turn to the control of another correlated tunneling process, i.e., ac-driven superexchange. The superexchange of particles mediated via single-particle tunneling to off-resonant intermediate states is the basic next-neighbor interaction mechanism in models of quantum magnetism arising in two-component Mott insulators [8,19,20]. In the presence of a magnetic-field gradient, the exchange of a pair of opposite spins $|\uparrow,\downarrow\rangle \rightarrow |\downarrow,\uparrow\rangle$ on a lattice is associated with an energy cost of $2G$ and superexchange processes are thus inhibited as soon as $G \gg J_{ex}^0$, where $J_{ex}^0 = 4J^2 / U$ is the exchange coupling in the absence of the gradient. By modulating the lattice potential at the resonance condition $\hbar\omega = 2G$, however, it is possible to restore the resonant exchange of spins.

As in the undriven case [8], the system can in the dressed state picture be mapped onto an effective XXZ model within second order of the tunnel couplings [21]

$$\hat{H}_{eff} = -\sum_{\langle i,j \rangle} [J_{ex}^0(\hat{S}_i^z \hat{S}_j^z + \hat{S}_i^y \hat{S}_j^y) + J_{ex}^0 \hat{S}_i^z \hat{S}_j^z]. \quad (2)$$

Hereby, the exchange of two particles in the presence of the driving can happen via four different intermediate states which are accessed via tunnel coupling with the amplitudes $J$ or $K$ [see Fig. 3(a)]. This gives rise to an effective transverse coupling on resonance of

$$J_{ex}^t = 2JK\left[\frac{1}{U + \Delta + G} + \frac{1}{U + \Delta - G} \right] + \frac{1}{U - \Delta + G} + \frac{1}{U - \Delta - G}. \quad (3)$$

where we include the possibility to apply an alternating energy offset $\Delta$. On the other hand, the energy shift due to virtual exchange processes which leave the spin configuration unchanged stems from couplings via six possible intermediate states and consists of terms $\propto J^2$ and $\propto K^2$ [21]. This difference allows for an arbitrary tuning of the anisotropy $J_{ex}^t / J_{ex}^0$. For example, in the case $J^2 / U \ll G \ll U$, $\Delta = 0$, and for small driving amplitude one obtains $J_{ex}^t \approx 8JK / U$ while the spin coupling in the $z$ direction is identical to the undriven case $J_{ex}^z \approx 4J^2 / U$.

The anisotropy thus is $\approx J / 2K$ and a weak driving ($K \ll J$) realizes an Ising-type model. Similarly, regimes can be found, where $J_{ex}^z$ vanishes [21].

In order to demonstrate the feasibility of the proposed method, we probe the transverse coupling $J_{ex}^t$ in our system of double wells [see Fig. 1(c)]. For this purpose, we loaded the atomic spin pairs into symmetric double wells in the presence of a magnetic-field gradient along the $x$ direction. Its strength $G / h = 1.2(1)$ kHz was measured from the shift of the single-particle resonance of $|\downarrow\rangle$ atoms. The degeneracy between $|\uparrow,\downarrow\rangle$ and $|\downarrow,\uparrow\rangle$ was hence lifted and the atoms were occupying the ground state $|\uparrow,\uparrow\rangle$. We carried out the modulation spectroscopy as in the previous cases, and a typical spectrum is displayed in Fig. 3(b) for $\Delta / h = 8.4(2)$ kHz. We observe two kinds of resonances. For $\omega = 2\pi / 4 = 5(2)$ kHz and $\omega = 2\pi / 13 = 3.0(2)$ kHz only the atoms in one of the spin states are transferred. For the second type of resonances both spin states are transferred simultaneously in an ac-driven superexchange process.

| FIG. 3 (color). (a) Sketch of the resonantly driven superexchange coupling via four possible intermediate states with different detunings $U \pm \Delta \pm G$, reached either by bare ($J$, solid arrows) or driven tunneling ($K$, dashed arrows). (b) Fraction of atoms $n_{\uparrow,\downarrow}$ in the right well as a function of the modulation frequency $\omega / 2\pi$. The lattice parameters were $V_x = 192(6)E_r$, $V_y = 142(5)E_r$, $V_0^p = 35(1)E_r$, $V_0^s = 7.0(2)E_r$, $\phi = 0.35(1)$ rad, and $G / h = 1.2(1)$ kHz. The simultaneous transfer of both spins for $\omega / 2\pi \approx 2.6$ kHz (gray line) corresponds to a single-photon-driven superexchange, while the one for $\omega / 2\pi \approx 1.5$ kHz is driven by the absorption of two photons. (c) Time evolution of the mean population imbalance $X(t)$ and spin imbalance $N^s(t)$ at the superexchange resonance $\omega / 2\pi = 2.6$ kHz, fitted with a damped sine (solid line). The fit yields a superexchange coupling of $J_{ex}^t / h = 0.56(2)$ kHz and a damping time $(1/\epsilon) = 9(1)$ ms.

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We now focus on the resonance occurring at $\omega/2\pi = 2.6(1)$ kHz, which corresponds to the resonance condition $\hbar\omega \approx 2G$ for the driven superexchange. As shown by the Rabi oscillation in Fig. 3(c), on resonance both spin states coherently tunnel between left and right wells in a correlated manner. At any time the population imbalance between both wells $X(t) = (n_L - n_R)/2$ remains equal to 0, as expected for a spin-exchange process. The actual states involved in the superexchange process are not fully localized due to the finite tunnel coupling $J$ and the coupling to higher bands. We calculate that for our trap parameters the maximum value of the mean spin imbalance $N^z = (n_{1L} - n_{1R} - n_{2L} + n_{2R})/2$ amounts to 0.8, close to the measured value $N^z(t = 0) = 0.7$. The damping most likely stems from inhomogeneities in $\Delta$ and $G$. Because of the latter, the Rabi oscillation is detuned for part of the atomic sample, leading to the observed nonzero asymptotic value.

In Fig. 4, we show a measurement of the superexchange coupling strength $|J_{ex}|$ as a function of the tilt $\Delta$. We observe a resonant enhancement of $|J_{ex}|$ over a factor of 5 when decreasing $\Delta$ from 3.9 to 2 kHz, approaching $\Delta = U - G = \hbar \times 1.7$ kHz, where the intermediate state $|L\rangle; 0; \nu - 1$ comes into resonance [see Fig. 3(a)]. The second-order perturbation theory within the two lowest Bloch bands, Eq. (3), gives a qualitative description of our measurements (dotted line), but breaks down near such resonances. We obtain a better quantitative agreement by including virtual transitions to higher bands (solid line). The remaining discrepancy can be attributed to system inhomogeneities, i.e., larger tunnel couplings $J$ and $K$ at the borders of the atomic sample.

To conclude, we have shown that periodically modulated optical superlattices provide a new tool for controlling correlated tunneling processes such as cotunneling and superexchange. Extending the concept of driven superexchange to a full lattice provides a novel approach to implement effective-spin XXZ models with arbitrarily tunable couplings and coupling anisotropy without the need to adjust the individual interaction energies. Both the measured coupling strength and the decoherence of driven superexchange oscillations are comparable with the ones of bare superexchange [20], indicating, in particular, that the lattice modulation would not add significant heating when generalized to a full lattice. It therefore opens the way to, e.g., experimentally study the relaxation of an initially Néel-ordered state in one [22], two, or three dimensions over a wide range of models ranging from XY-type over Heisenberg-type to Ising-type. It is possible to combine the technique with proposed schemes for preparing a fermionic antiferromagnet from a low entropy band-insulating initial state [23]. However, their experimental realization remains challenging.

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Note added.—Recently, we became aware of related work by Ma et al. [25].

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*Yu-Ao.Chen@lnu.de

[24] All lattice depths are given in units of the respective recoil energy $E_i^r = h^2/2m\lambda_i^2$, where $i = xx, xl, y, z$.