Giant Resonant Self-Blocking in Magnetophononically Driven Quantum Magnets

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Magnetophononics, the modulation of magnetic interactions by driven infrared-active lattice excitations, is emerging as a key mechanism for the ultrafast dynamical control of both semiclassical and quantum spin systems by coherent light. We demonstrate that, in a quantum magnetic material with strong spin-phonon coupling, resonances between the driven phonon and the spin-excitation frequencies exhibit a giant self-blocking effect. Instead of absorbing more energy, the spin system acts as a strong brake on the driven phonon, causing it to absorb only a tiny fraction of the power available from the laser. Using the quantum master equations governing the nonequilibrium steady states of the coupled spin-lattice system, we show how self-blocking dominates the dynamics, demonstrate the creation of mutually repelling hybrid spin-phonon states, and control the nonequilibrium renormalization of the lattice-driven spin excitation band.

Rapid advances in laser technology [1] have made it possible not only to probe but also to pump quantum materials in a controlled manner on ultrafast timescales and at all the frequencies relevant to excitations in condensed matter [2–4]. This has led to phenomena ranging from Floquet engineering of electronic band structures [5] to enhanced superconductivity [6] and switching of the metal-insulator transition [7]. A wide range of experimental and theoretical efforts is now under way to extend such ultrafast control to every aspect of strongly correlated materials beyond the charge, including lattice, orbital, spin, nematic, and chiral degrees of freedom [8].

Among these, spin systems offer perhaps the ultimate quantum many-body states due to their intrinsically high entanglement and relatively low energy scales, which lead to rather clean experimental realizations. Ultrafast switching, modulation, transport, and destruction of semiclassical ordered magnetism have been achieved using light of different frequencies [9] [11]. However, coupling to a magnetic order parameter is often not appropriate for the dynamical control of quantum magnetic materials, and increasing attention is focused on using the lattice as an intermediary [12–16]. While “nonlinear phononics” [17] exploits the anharmonic lattice potential, to date for low-frequency magnetic control [18]. “magnetophononics” [19] uses harmonic phonons to gain fully frequency-selective control of exchange-type interactions [20]. Strong excitation of collective spin modes at their intrinsic frequencies opens the possibility not only of effecting the slow or fast (Floquet) modulation of existing magnetic states but also of creating fundamentally different types of hybrid state.

In this Letter we show that the magnetophononic mechanism has an intrinsic giant self-blocking effect, by which a driven phonon in resonance with the peak density of magnetic excitations absorbs very little of the driving laser power. We demonstrate self-blocking by considering the nonequilibrium steady states (NESS) of an alternating quantum spin chain strongly coupled to a bulk Einstein phonon mode. We compute the driving-induced mutual renormalization of the lattice and spin excitations, finding that distinctive hybrid excitations emerge for phonon frequencies near the spin-band edges and that other in-band frequencies effect a global reshaping of the spin spectrum. We discuss the consequences of self-blocking and dynamical spectral renormalization for pump-probe experiments on quantum magnetic materials such as CuGeO$_3$ and (VO)$_2$P$_2$O$_7$.

To analyze the dynamics of a phonon-driven and dissipative quantum magnet, we use the alternating $S = 1/2$ spin chain discussed in Ref. [21] and depicted in Fig. 1a. A bulk Einstein phonon, which is infrared (IR)-active to be excited coherently by the incident light, modulates one of the magnetic superexchange interactions at the driving frequency. The elementary excitations of the chain are gapped triplons, and the band of two-triplon modes excited by the driven phonon sets the resonant frequencies of the coupled system. Both this phonon and the spin excitations have a lifetime, i.e. a damping, due to all the other (acoustic and optic) phonons of the lattice.

We treat this open quantum system by introducing Lindblad operators [22], the operators of the isolated system describing its interaction with the damping “bath,” to deduce the equations of motion governing the time evolution of its physical observables. A complete derivation may be found in Ref. [21] and, for a self-contained presentation, is summarized in Sec. S1 of the Supplemental Material (SM) [23]. Here we state only our primary assumptions, that the Lindblad operators for the Einstein phonon are its own creation and annihilation operators, respectively $b_0^\dagger$ and $b_0$, while those for the spin sector are the triplon creation and annihilation operators, $\tilde{t}_{k\alpha}^\dagger$ and $\tilde{t}_{k\alpha}$, for each reciprocal-space mode of an $N$-dimer spin chain. These assumptions, whose physical meaning is also described in Sec. S1, ensure a straightforward system of $3(N/2 + 1)$ equations of motion with no mixing
between the triplon modes at different values of \( k \).

In the NESS established by steady laser driving, the phonon occupation, \( n_{ph}(\omega) \equiv \langle \hat{n}_{\omega} \rangle \), is approximately constant for all \( \omega \), and the density of two-triplon excitations is highest. For \( J'/J = 0.5 \) as in Fig. 1(b), the edges of the two-triplon band lie at \( 2\omega_{\text{min}} = 1.444 J \) and \( 2\omega_{\text{max}} = 2.449 J \), and this resonant effect becomes gigantic at strong \( g \), suppressing the phonon occupation by nearly three orders of magnitude at \( 2\omega_{\text{min}} \).

We have named this effect “self-blocking” because the magnetic system acts to block its own energy uptake by blocking the driven phonon. This behavior is surprising if one expects stronger energy absorption when more spin excitations coincide with the driving laser frequency. Its explanation lies in the fact that in magneto-phononically driving the spin system is not coupled to the light, but only to the driven phonon. Heuristically, it acts as an extra “inertia” for the phonon to drive into motion. Analytically, the prefactor of the phonon momentum, \( p(\Omega) = \langle 1/\gamma_\omega (b_0^\dagger - b_0) \rangle(\Omega) \), in the master equation for \( n_{ph}(\omega) \) contains terms by which the spin system acts directly against the driving electric field, suppressing its effective value to \( E_\Omega(\omega) = E_\Omega(\omega) - g \sum_k \langle f(t_k, \omega) \rangle \) (Secs. S1 and S2 of the SM [23]). This negative feedback effect is strongly nonlinear in \( g \) and can cancel \( E_\Omega(\omega) \) almost completely at resonance [Fig. 1(b)]. Despite the approximate symmetry of the spin band, self-blocking is weaker by a factor of 10 at \( 2\omega_{\text{max}} \) due to matrix elements within the feedback process.

Away from the two-triplon band, in Fig. 1(b) we observe a significant suppression of phonon energy entering the system at any frequency \( \omega_0 < 2\omega_{\text{min}} \). This nonresonant self-blocking is also nonlinear in \( g \), exceeding one order of magnitude at \( g = 0.5 J \). Its appearance only in the low-\( \omega_0 \) regime, but not at \( \omega_0 > 2\omega_{\text{max}} \), points to an origin in multiple harmonic processes \( 2\omega_{\text{min}} < n\omega_0 < 2\omega_{\text{max}} \) [21]. Although only the two-phonon harmonic \( (n = 2) \) at \( \omega_{\text{min}} \) is present directly, stronger \( g \) distributes the response of the system to two broader ranges of frequencies. By contrast, a driving phonon at the band center \( (\omega_0 = 0.2 J) \) has vanishing matrix elements with the resonant spin modes, and hence \( n_{ph0} \) recovers almost to its \( g = 0 \) value for all \( g \).

Turning to the response of the spin system, Fig. 1(c) shows the corresponding average triplon occupancy, \( \bar{n}_{x0} \). The most striking feature is the strong rounding of the in-band response as \( g \) is increased. The band-edge peaks are entirely blunted by the strong suppression of \( n_{ph0} \) [Fig. 1(b)]. We stress that the effective limiting value \( \bar{n}_{x0} \approx 0.1 \) visible in Fig. 1(c) is purely a consequence of the giant self-blocking, and is not connected with the hard-core nature of the triplon excitations, which has not been included in our formalism [21]. This rounding suggests an increasing localization of the spin response, by which the band character of the triplons becomes less relevant under strong driving by the entirely local phonon. In Figs. 1(b) and 1(c) the system is driven at the phonon frequency, \( \omega_0 \). It is important to note that self-blocking is not a simple shift of phonon energy to different frequencies: NESS established by steady driving contain no frequencies other than those of the drive and its higher harmonics [23]. To probe how a strong \( g \) modifies

![Diagram](image-url)
FIG. 2. Strongly and weakly hybridized excitations. (a) Phonon occupation, \( n_{\text{ph}} \), shown as a function of driving frequency, \( \omega \), for a phonon frequency \( \omega_0 = 1.35 J \) at selected \( g \) values. The standard driving and damping parameters of Fig. 1(b) are used. (b) As in panel (a) for \( \omega_0 = 1.45 J \). (c) \( n_{\text{ph}}(\omega) \) (dotted lines) and triplon occupation, \( n_{\text{t}}(\omega) \) (solid lines), shown for a phonon frequency, \( \omega_0 = 0.6 J \), far from the spin excitation band. (d) \( k \)-resolved components of the average \( \langle u_k(\omega) \rangle \) at \( \omega_0 = 0.6 J \). (e) \( n_{\text{ph}}(\omega) \) for \( \omega_0 = 1.35 J \), corresponding to panel (a). (f) \( n_{\text{t}}(\omega) \) for \( \omega_0 = 1.45 J \), corresponding to panel (b). (g) Peak-pair frequencies, labelled \( \omega_1^{\text{hyb}} \) and \( \omega_2^{\text{hyb}} \), as in panels (a), (b), (e), and (f), shown for all \( g \) values; black dashed lines indicate a \( g^2 \) form. (h) Data of panel (b) normalized to the resonant phonon occupation, \( \pi_{\text{ph}}(\omega) \), at the same \( \omega \) [data of Fig. 1(b)].

the spin and phonon spectra, we begin with the phononic response of a system driven at frequencies \( \omega \neq \omega_0 \). Focusing first on \( \omega_0 \) values near resonance, Fig. 2(a) shows a phononic excitation that lies just below the two-triplon band being weakened and pushed away from the lower band edge at stronger \( g \), i.e. a level repulsion. Figure 2(b) shows the analogous result when \( \omega_0 \) lies just inside the spin band, where the phonon peak is damped very strongly with increasing \( g \), and is also repelled from the band edge. Here it is accompanied by the development of a second feature, appearing at \( 2\omega_{\text{min}} \) at \( g = 0.1 J \), which is repelled below the band edge as \( g \) increases.

Before proceeding, the giant self-blocking at resonance raises the question of whether this two-peak effect could be just a secondary consequence of the very strongly suppressed phononic response at \( \omega = 2\omega_{\text{min}} \). For a heuristic measure of self-blocking we show in Fig. 2(h) the result of Fig. 2(b) normalized by \( \pi_{\text{ph}} \) from Fig. 1(b). The weak-\( g \) peaks then appear with unit magnitude, while the strong-\( g \) phononic response does confirm a two-peak structure with level repulsion, the induction of hybrid spin-phonon states. Here the in-band hybrid remains damped, whereas the hybrid generated outside the spin band responds much more strongly per unit driving.

To confirm the hybrid nature of these states we examine their spin character. When \( \omega_0 \) is far from the two-triplon band, \( n_{\text{t}}(\omega) \) indicates that a magnetic response emerges with \( g \) despite the nonresonant self-blocking [Fig. 2(c)]. The minor changes in \( n_{\text{ph}}(\omega) \) indicate that this is a localized phononic mode whose weak hybridization is enough to shift its frequency out of Fig. 1(c) and whose dressing at \( g = 0.5 J \) involves all the \( k \)-components of \( n_{\text{t}}(\omega) = \sum_k u_k(\omega) (u_k = \sum_\alpha (f_{k\alpha}^t f_{k\alpha})) \) [Fig. 2(d)]. Returning to the band edges, Figs. 2(c) and 2(f) complement Figs. 2(a) and 2(b), and in Fig. 2(g) we gather the characteristic frequencies of these phonon and spin spectra [including the second peak weakly visible inside the band in Fig. 2(a)], highlighted on a logarithmic scale in Sec. S2 of the SM [23]. These display the clear development with \( g \) of two mutually repelling hybrid excitations whose frequency shifts scale accurately with \( g^2 \); we show in Sec. S3 that the same physics is also found for driving frequencies around the upper band edge.

Concerning the admixture of lattice and spin character, if one defines a hybridization parameter \( s = g/|\omega_0 - 2\omega_{\text{min}}| \), then a language of “phononic” and “magnetic” hybrids remains useful at \( \omega_0 = 0.6 J \) [Fig. 2(c)], where \( s < 1 \) for all \( g \). However, when \( s \approx 10 \) both hybrids are strongly magnetic and phononic, and indeed the 50:50 weight distribution at larger \( g \) in Figs. 2(c) and 2(f) suggests states that are maximally hybridized. For the hybrids repelled outside the band, the coinciding peaks in \( n_{\text{ph}}(\omega) \) and \( n_{\text{t}}(\omega) \) identify them as a strongly triplon-dressed version of the “phononic” hybrid shown in Figs. 2(c) and 2(d). The in-band hybrids lie in a continuum of propagating triplon-pair states, and thus manifest themselves as broader peaks, lying at slightly different energies, in \( n_{\text{ph}}(\omega) \) [Fig. 2(b)] and \( n_{\text{t}}(\omega) \) [Fig. 2(f)].
For the driving and damping of our system, all hybrid states are to a good approximation "phonon-bitriplons," i.e. phonons dressed by one triplon pair \((\not{t}_0 \not{t}_0^+)\) of zero net momentum. Despite the ubiquity in physics of "light-matter" interaction processes where a boson produces pairs of fermions, processes where one boson produces pairs of bosons are rare. Phonon-bimagnon processes have been discussed both theoretically [24, 25] and experimentally [26, 27] in the optical spectra of cuprate quantum magnets. Similar physics could be engineered using ultracold atoms [28], where the optical lattice blurs the distinction between photon and phonon, although we are not aware of an experiment. On a very different energy scale, in particle physics the virtual decay of the Higgs boson into pairs of W or Z bosons [29, 30] is an off-shell process with intermediate \(s\), where the level repulsion of Fig. 2(g) is known as a "Higgs mass renormalization."

Having demonstrated both strong driving-induced changes to the excitation spectra and strong suppression of this driving near resonance, we consider the consequences for the properties of the NESS. Figure 3(a) shows that \(\bar{q}_0\), the average of the phonon displacement \([q(t) = \langle \frac{1}{\sqrt{N}} (\not{b}_0 + \not{b}_0^+) \rangle(t)\] obtained when driving at \(\omega = \omega_0\), is pushed to a finite value, which can reach 4% of the lattice dimension at \(g = 0.5J\). In our minimal model, this driven distortion implies a linear modification of the magnetic interaction \(J\) to \(J = J + g \bar{q}_0\). The associated renormalization of the two-triplon band is partly reflected in the evolution of \(u_{k=\pi}(\omega)\) on increasing \(g\), as we show in Sec. S4 of the SM [23]. However, for an accurate measurement we introduce the "pump-probe" protocol of Fig. 1(a), driving the system at \(\omega = \omega_0\) and monitoring its response to an additional, weak probe component of the laser at frequency \(\Omega\). Because we are investigating NESS, \(E(t) = E_0 \cos(\omega t) + E_1 \cos(\Omega t)\) is continuous and the time delay used in true pump-probe studies is absent.

As the most sensitive diagnostic of the edges of the modified two-triplon band, in Figs. 3(b) and 3(c) we show not \(n_{k=\pi}(\omega_0, \Omega)\) but the respective components \(u_{k=\pi,0}\) and \(u_{k=\pi,\omega}\). As \(\Omega\) is scanned through the band-edge frequencies, the peaks in both quantities grow and shift away from the equilibrium band edges when \(g = 0.3J\) and \(0.5J\). In contrast to Figs. 2(e) and 2(f), where two strongly hybridized excitations form at the band edge due to the near-resonant phonon mode, \(s < 1\) when the system is driven at \(\omega = \omega_0 = 2.2J\) and we observe a single peak in the magnetic response. When corrected for the corresponding weak hybridization shift, \(\delta \omega_s < 0.005J\), which we show in Sec. S4 of the SM [23], these peaks indicate a driving-induced renormalization of the entire two-triplon band. Assuming that only \(J\) is renormalized yields quantitative agreement with the peaks observed in our pump-probe spectra, i.e. the triplon band retains its cosine form (also shown in Sec. S4) in the model of Fig. 1(a).

This dynamical renormalization is again a strongly nonlinear function of \(g\), and demonstrates how magnetophonic driving can be used to control the spin states at frequencies far from that of the pump. The key physics contained in Fig. 3(a) is that the phonon frequencies most effective for exerting this control are neither those at the band edges, where giant self-blocking [Fig. 1(b)] suppresses \(\bar{q}_0\) almost completely, nor those at the band center, where the driving terms decouple, but the "quarter-band" ones around \(k = \pi/4\) and \(3\pi/4\). Considering a model with the phonon coupled instead to the \(J^*\) bond in Fig. 1(a) reinforces these conclusions while allowing a different type of band renormalization, as we show in Sec. S5 of the SM [23]. Although the band shifts in Figs. 3(b) and 3(c) are not large for our standard driving (solid lines), doubling the electric field leads to a very strong effect (dotted lines), such that a majority of the total weight in the nonequilibrium (driven) spin spectrum can appear at frequencies that are forbidden at equilibrium.

Turning to experiment, CuGeO$_3$ [31] and (VO)$_2$P$_2$O$_7$ [32] are quantum magnetic materials known to have very large \(g\). Both have low structural symmetry, making IR-active phonons available over a range of energies around relatively broad spin bands [33, 34]. Experiments to cre-
ate spin NESS at the resonant frequencies of bulk-driven quantum magnets require a thin-film geometry and efficient thermal transfer to maintain a low sample temperature [21]. In principle, self-blocking can allow significant relaxation of the constraints on pump intensity, driving time, and sample thickness, although in practice strong electromagnetic driving can induce heating by a variety of channels. For controlling the spin excitation spectrum, we comment that $\tilde{J}(q_0)$ is in general a highly nonlinear function, and although conventional experimental probes usually require only perturbative expansions, coherent laser driving can produce very large $q_0$ values [20].

Self-blocking is favored by a high density of spin states. Thus it should be prevalent not only at the band edges in low-dimensional quantum magnets but also in any system with nearly flat spin bands, which can arise in strongly correlated materials of any dimensionality. SrCu$_2$(BO$_3$)$_2$ is one such system [37], in which a recent experiment has demonstrated a phonon-driven nonequilibrium population of two-triplon excitations [20], but not yet a self-blocking. We stress also that the dissipative processes in our analysis are generic and hence the phenomenology of self-blocking is independent of the nature of the bath, as discussed in Sec. S1 of the SM [23].

Coherent phononic driving can be applied in quantum magnetic materials at high phonon frequencies to Floquet-engineer collective spin states and with slow phonons to modulate the existing magnetic energy levels. Resonant magnetophononic driving goes beyond these situations by creating qualitatively different hybrid quantum states and feedback effects. In this regime we have discovered an initially counterintuitive giant self-blocking. We explain this phenomenon and place it in the context of controlling the dynamical renormalization of spin states by light through the medium of the lattice.

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Supplemental Material for “Giant Resonant Self-Blocking in Magnetophononically Driven Quantum Magnets”

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S1. QUANTUM MASTER EQUATIONS

A. Coupled phonon and spin systems

We consider a minimal Hamiltonian describing a straightforward and well understood quantum spin system coupled strongly to a single, nondispersive optical phonon. A detailed derivation of the appropriate diagonal Hamiltonians and the equations of motion governing the physical observables is presented in Ref. [24]. This subsection provides a summary of the methods, notation, and underlying physics.

The quantum spin system is an alternating $S = 1/2$ Heisenberg chain, which can be described in terms of the operators $\hat{t}_{k\alpha}$ and $\hat{E}_{k\alpha}$ creating and destroying triplon excitations [Fig. 1(a) of the main text]. The spin Hamiltonian in reciprocal space takes the form

$$H_s = \sum_{k,\alpha} \omega_k \hat{t}_{k\alpha}^{\dagger} \hat{t}_{k\alpha}, \quad (S1)$$

with the dispersion relation

$$\omega_k = J \sqrt{1 - \lambda \cos k}, \quad (S2)$$

where $\lambda = J'/J$.

For illustration we consider an Einstein phonon mode coupling strongly to only one of the magnetic interactions of the spin system and driven by the electric field of the laser. The relevant Hamiltonian terms in real space are

$$H_p + H_{sp} + H_1 = \sum_{j} \left[ \omega_0 b_j^\dagger b_j + g(b_j + b_j^\dagger) \hat{S}_{1,j} \cdot \hat{S}_{2,j} + E(t)(b_j + b_j^\dagger) \right], \quad (S3)$$
where $\tilde{S}_{1,j}$ and $\tilde{S}_{2,j}$ are the $S = 1/2$ spin operators on dimer $j$ of the chain, $g$ is the spin-phonon coupling constant, and $E(t) = Eq \cos(\omega t)$ is the single-frequency oscillating electric field of the light. To describe the physical observables of the driven phonon we consider the real variables

\[ q(t) = \langle \frac{1}{N} (b_0 + b_0^\dagger) \rangle(t), \quad (S4a) \]
\[ p(t) = \langle \frac{1}{N} (b_0^\dagger - b_0) \rangle(t), \quad (S4b) \]
\[ n_{\text{ph}}(t) = \langle \frac{1}{N} b_0^\dagger b_0 \rangle(t), \quad (S4c) \]

at any given time, $t$, which correspond respectively to the displacement of the Einstein phonon, the conjugate momentum, and the number operator.

To formulate analogous quantities for the spin sector we consider the $k$-space components of the triplon number operator and its off-diagonal equivalent,

\[ u_k = \sum_\alpha \tilde{t}_{\alpha k}^\dagger \tilde{t}_{\alpha k} \text{ and} \quad (S5a) \]
\[ \tilde{v}_k = \sum_\alpha \tilde{t}_{\alpha k}^\dagger \tilde{t}_{-\alpha k}. \quad (S5b) \]

We denote their expectation values by

\[ u_k(t) = \langle u_k(t) \rangle, \quad (S6a) \]
\[ \tilde{v}_k(t) = \langle \tilde{v}_k(t) \rangle, \quad (S6b) \]

and similarly for $\tilde{v}_k^\dagger(t)$, then separate the real and imaginary parts to obtain

\[ u_k(t) = \Re \tilde{v}_k(t) \quad (S7a) \]
\[ w_k(t) = \Im \tilde{v}_k(t). \quad (S7b) \]

$u_k(t)$ is a real variable and a valuable characterization of the driven spin sector is provided by the nonequilibrium triplon number,

\[ n_x(t) = \frac{1}{N} \sum_k u_k(t). \quad (S8) \]

It is convenient to define the coefficients

\[ y_k = \frac{1 - \frac{1}{2} \lambda \cos k}{\sqrt{1 - \lambda \cos k}} = \frac{J 1 + \omega_k^2 / J^2}{\omega_k} \quad (S9a) \]
\[ y_k' = \frac{\frac{1}{2} \lambda \cos k}{\sqrt{1 - \lambda \cos k}} = \frac{J 1 - \omega_k^2 / J^2}{\omega_k}, \quad (S9b) \]

and the real quantities

\[ U(t) = \frac{1}{N} \sum_k y_k [u_k(t) - 3n(\omega_k)] \quad \text{and} \quad (S10a) \]
\[ V(t) = \frac{1}{N} \sum_k y'_k w_k(t), \quad (S10b) \]

where $n(\omega_k) = [\exp(\hbar \omega_k / k_B T) - 1]^{-1}$, the bosonic occupation function for the triplon mode with frequency $\omega_k$, is an accurate approximation to the true occupancy of the hard-core triplon modes for small $n_{\text{ph}}$.

**B. Equations of motion**

The time evolution of an open quantum system is specified by adjoint quantum master equations of the form

\[ \frac{d}{dt} A_H(t) = i[H, A_H(t)] \quad (S11) \]
\[ + \sum_i \tilde{\gamma}_i [A_i^\dagger A_H(t) A_i - \frac{1}{2} A_H(t) A_i^\dagger A_i - \frac{1}{2} A_i A_i^\dagger A_H(t)] \]

for any operator $A_H(t)$ describing a physical observable. In these Heisenberg equations of motion, $H$ is the Hamiltonian of the isolated quantum system, excluding the bath. The Lindblad operators, $\{A_i\}$, are also formed from those of the isolated system and the coefficients $\tilde{\gamma}_i$ are effective damping parameters. The Lindblad operators for the driven phonon are $A_1 = b_0^\dagger$ and $A_2 = b_0$, with corresponding damping coefficients $\gamma_1 = \gamma n(\omega_0)$ and $\gamma_2 = \gamma [1 + n(\omega_0)]$, which return the equations of motion of the damped harmonic oscillator,

\[ \frac{d}{dt} q(t) = \omega_0 p(t) - \frac{1}{2}\gamma q(t), \quad (S12a) \]
\[ \frac{d}{dt} p(t) = -\omega_0 q(t) - \frac{1}{2}\gamma p(t) - 2E(t), \quad (S12b) \]
\[ \frac{d}{dt} n_{\text{ph}}(t) = -\tilde{E}(t) p(t) - \gamma [n_{\text{ph}}(t) - n(\omega_0)], \quad (S12c) \]

in which $E(t) = E(t) + g(U(t) + V(t))$ expresses the effective electric field of the light acting on the Einstein phonon in the presence of renormalization arising from the spin system to which the phonon is coupled.

Turning to the spin system, we adopt as the Lindblad operators the linear one-triplon operators, $\tilde{t}_k$ and $\tilde{t}_k^\dagger$, with a single spin-damping coefficient, $\tilde{\gamma}_k = \gamma n(\omega_k)$. As discussed in Ref. [21], these are not spin-conserving operators: physically, their meaning is that a phonon oscillation can change the spin quantum number and thus they are appropriate for systems with appreciable spin-orbit coupling. In a real material, their effects would be accompanied by those of bilinear Lindblad operators (of the form $C_{ka} = \tilde{t}_k^\dagger \tilde{t}_a$), which would be the leading bath terms in a system with weak spin anisotropy. However, here we use only the linear operators for simplicity of presentation and for physical transparency. The equations of motion of the spin sector are then

\[ \frac{d}{dt} u_k(t) = 2gq(t) y'_k w_k(t) - \gamma_3 [u_k(t) - 3n(\omega_k)], \quad (S13a) \]
\[ \frac{d}{dt} v_k(t) = -2[\omega_k + g y_k q(t)] w_k(t) - \gamma_4 v_k(t), \quad (S13b) \]
\[ \frac{d}{dt} w_k(t) = 2[\omega_k + g y_k q(t)] v_k(t) \]
\[ + 2gq(t) y'_k [u_k(t) + \frac{3}{2}] - \gamma_5 w_k(t), \quad (S13c) \]

for each mode $k$ of the spin chain.

We solve the $3(N/2 + 1)$ equations of motion for spin chains with lengths up to $N = 2000$ dimers. We focus on
the nonequilibrium steady states (NESS) of the driven, dissipative system, whose formation at small $g$ values requires a timescale of approximately $5/\gamma_s$, i.e. five time constants of the spin system [21]. However, NESS formation at large $g$ values may require several cycles through strong feedback processes, and to deal with these cases we consider times up to $t = 30000/J$. An example of complementary phonon and spin NESS, each characterized by their number operators, is shown in the time domain for a nonresonant system frequency, $\omega_0 = \omega_{\min}$, in Fig. S1 at weak, strong, and very strong values of $g$. We observe that both time traces contain increasingly complex combinations of harmonics as $g$ is increased; the amplitude of the oscillatory part of the phonon occupation starts to be suppressed at very strong $g$; the oscillatory part of the triplon occupation rises very strongly with $g$ on exiting the weak-coupling regime, but is also suppressed at very strong coupling. The corresponding static parts of the both occupations, $\pi_{ph0}$ and $\pi_{s0}$, may be read respectively from Figs. 1(b) and 1(c) of the main text for all phonon frequencies.

Concerning the timescale for the development of self-blocking, Fig. S1(c) shows how the initial rise of $n_{ph}$ is truncated by the rise of $n_x$ [Fig. S1(d)]. In this nonresonant regime, at $g = 0.3J$ there remains a significant time lag between the driving phonon and following triplon occupations, where the latter limits the former and convergence requires one cycle. At $g = 0.5J$, the lag in response is much shorter and several slow oscillation cycles are required. The same phenomena are visible at $\omega_0 = 2\omega_{\min}$ (not shown), except that the resonant self-blocking is so extreme that it is difficult to see the $n_{ph}(t)$ curves for all $g$ on the same scale, and convergence can require more than 5 slow oscillation cycles.

We comment that the equations of motion are valid at all times from the onset of driving ($t = 0$) to infinity and for all applied electric fields, as well as for all phonon occupations up to the Lindemann melting criterion ($n_{ph} \approx 3$). With the present simplified treatment of the spin sector, they are valid up to a triplon occupation of order $n_x \approx 0.2$, beyond which a more sophisticated numerical treatment should be used to account for the hard-core occupation constraint. Because the equations of motion are based on a mean-field decoupling of the spin and lattice sectors, our treatment becomes more approximate at low phonon frequencies [21], specifically those below $\omega_0 = 0.2-0.3J$ on the left side of Fig. 1 of the main text. Nevertheless, one may verify by considering the energy flow through the strongly spin-phonon-coupled system that the mean-field approximation remains very accurate at resonance ($\omega_0 = 2\omega_{\min}$), i.e. that its deterioration is a consequence of the frequency range, not only of a very low energy flux within the system, and thus that it is not a factor in self-blocking.

Finally, one may question the stability of the alternating chain in the presence of phononic driving, particularly when this is very strong or very slow. While self-blocking limits the effectiveness of strong driving, in fact the sharp fall in $\pi_{ph0}$ at very small $\omega_0$ in Fig. 1(b) of the main text is related to a ground-state instability of the chain, where a stimulated distortion can occur ($q_0$ becomes finite) in the presence of sufficiently slow phonons. One may show that the stability criterion takes the form $\omega_0^2 > F(\lambda)g^2\lambda^2J$, and that for $\lambda = 0.5 = g/J$ this critical

![Figure S1](image-url)
value is $\omega_0^* \approx 0.07J$.

S2. SELF-BLOCKING

Here we provide two sets of comments augmenting the discussion of self-blocking presented in the main text. Extra insight into the phenomenon is provided if, as in Fig. 2(h) of the main text, one considers each of the response functions of the spin chain normalized by the actual driving strength at the same frequency, which can be gauged from $n_{ph0}$. In Fig. S2 we show the normalized spin response, $\pi_{x0}$, of Fig. 1(c) of main text. This clearly regains the form of a weak-$g$ response at all $g$, with very strong peaks restored at $2\omega_{min}$ and $2\omega_{max}$ and the same relative intensities at all frequencies. Thus one may state that self-blocking is the only significant effect on the relative intensities, and beyond it one observes only a minor occupation at the midband energies as strong $g$ spreads the resonant response to a wider range of frequencies. It goes without saying that this fictitious spin response would be far beyond the physics of the spin chain, because of the hard-core nature of the triplon excitations, and a different physical mechanism would certainly limit the system to maximally nonequilibrium mode populations below $n_{ph0} = 1$, but self-blocking preempts this by providing a stricter limit.

As noted in the main text, it is also necessary to consider whether the self-blocking phenomenon could be a consequence of any simplifying assumptions made about the system or the bath. The self-blocking we observe is predominantly an effect of feedback on the Einstein phonon, whose coupling to the spin system is entirely conventional. Although we illustrated self-blocking for a simplified spin bath preserving the independent $k$ states of the triplons, even in this situation we demonstrated that strong spin-phonon coupling leads to strong shifts of spectral response in energy, and hence between modes at different $k$ values. Thus the phenomenon is by no means restricted to a single $k$ state at the resonant energy, and one may even speculate that a more efficient distribution of spectral weight among states of different $k$ could lead to more spin-damping contributions (the $\gamma_s$ terms of more active modes) and stronger self-blocking.

Finally, we comment that most ultrafast experiments to date use only very short driving pulses [8], which do not fit into the NESS framework we have investigated here, but as a consequence do not suffer the same heating problems [21]. They also use very strong electric fields, producing instantaneous atomic motion (and hence $n_{ph}$) rather than a dependence on the inverse damping times [$\gamma^{-1}$ and $\gamma_s^{-1}$ in Figs. S1(c) and S1(d)]. Under these circumstances one may anticipate that the spin-system feedback ($n_x$) also becomes instantaneous, and thus that the phenomenon of resonant self-blocking is equally applicable to an intensely driven phonon mode coupled strongly to a high density of spin states, although its nature in this transient time regime remains to be explored.

S3. HYBRID EXCITATIONS

We augment the analysis of the phonon and spin spectra shown in Fig. 2 of the main text by illustrating the ubiquitous nature of the spin-phonon hybridization phenomena. First, in Fig. S3(a) we provide the data of Fig. 2(a) of the main text showing $n_{ph0}(\omega)$ on a logarithmic scale. In this form it is clear that the “phononic” hybrid mode being repelled below the band edge with increasing $g$ is accompanied by a weak “magnetic” hybrid state forming inside the band (although we stress again that the figure shows the phononic character of this state). In Fig. S3(b) we present the analogous data for a phonon just above the upper band edge, at $\omega_0 = 2.5J$, which is the situation we discuss next.

Figure S4 shows the phononic and magnetic response of the system in the situation where the Einstein phonon frequency is close to the upper two-triplon band edge.
FIG. S4. (a) Phonon occupation, $n_{\text{ph}}(\omega)$, shown for a phonon frequency $\omega_0 = 2.4J$ at selected $g$ values. The standard driving and damping parameters of Fig. 1(b) of the main text are used. (b) As in panel (a) for $\omega_0 = 2.5J$. (c) $n_{\text{ph}}(\omega)$ (dotted lines) and triplon occupation, $n_{\text{xx}}(\omega)$ (solid lines), shown for a phonon frequency, $\omega_0 = 3.0J$, far from the spin excitation band. (d) $k$-resolved components of the average $u_k(\omega)$ at $\omega_0 = 3.0J$. (e) $n_{\text{xx}}(\omega)$ for $\omega_0 = 2.4J$, corresponding to panel (a). (f) $n_{\text{xx}}(\omega)$ for $\omega_0 = 2.5J$, corresponding to panel (b). (g) Peak-pair frequencies, labelled $\omega_{\text{hyb}}^1$ and $\omega_{\text{hyb}}^2$, as in panels (a), (b), (e), and (f), shown for all $g$ values; black dashed lines indicate a $g^2$ form. (h) Data of panel (a) normalized to the phonon occupation $\pi_{\text{ph}}$ at the same $\omega$ [data from Fig. 1(b) of the main text].

All of the features of Fig. S4 are at the qualitative level symmetrical with those observed in Fig. 2 of the main text, which showed the response when $\omega_0$ is close to the lower band edge. In Figs. S4(a) and S4(b) we show the development of mutually repelling hybrid states as gauged by their phononic response, $n_{\text{ph}}(\omega)$, in the presence of a phonon respectively just below or just above the band edge. The latter is the situation shown in Fig. S3(b), where in contrast to Fig. 2(a) of the main text the magnetic hybrid is in fact discernible (a result reflecting the differences in matrix elements governing the two situations). In Figs. S4(c) and S4(d) we show the weakly hybridized dressed phononic mode at $\omega_0 = 3.0J$, where the frequency shift due to hybridization is negligible and the weak spin response is dominated by $u_k = \pi_0$. In Figs. S4(e) and S4(f) we show the spin response, $n_{\text{xx}}(\omega)$, corresponding to Figs. S4(a) and S4(b), noting from the equal heights of the peaks on both sides of $2\omega_{\text{max}}$ at all large $g$ values that these are again close to the maximally hybridized situation. Figure S4(g) collects all of the characteristic frequencies of the phononic and magnetic facets of the strongly hybridized entities, which we denote by $\omega_{\text{hyb}}^1$ for the in-band peaks in $n_{\text{ph}}(\omega)$ and $n_{\text{xx}}(\omega)$, and by $\omega_{\text{hyb}}^2$ for the above-band peaks. As in Fig. 2(g) of the main text, outside (above) the band there is only one characteristic frequency for both types of response, whereas inside the band there is a not insignificant spread in peak values. Finally, in Fig. S4(h) we show the phononic response of Fig. S4(a) normalized by the self-blocking effect of Fig. 1(b) of the main text, which emphasizes again the very strong negative feedback effects operating for all phonon frequencies within the two-triplon band.

S4. DYNAMICAL RENORMALIZATION

To gain a more quantitative understanding of how the driving modifies the static properties of the coupled system, in Fig. S5(a) we show $\eta_0$ from Fig. 3(a) of the main text normalized by the actual phonon occupation, $\pi_{\text{ph}}$, taken from Fig. 1(b) of the main text. This allows one to gauge how the band renormalization caused by $\eta_0$ is in fact controlled completely by self-blocking, becoming very small at the band edges, precisely where one might have anticipated the strongest effects on the basis of Fig. S3(a).

In Figs. S5(c) and S5(d) we show how the band renormalization appears in the pump spectrum, when the system is driven at frequency $\omega$ with no probe signal. In this situation one may consider the components of $u_k$, whose average values peak strongly at the actual band frequency, defining a characteristic wave vector, $k_{\text{ren}}$ [21].

For a phonon at $\omega_0 = 1.45J$ [Fig. S5(c)], $u_{k_{\text{ren}}, 0}$ first undergoes a moderate enhancement due to the effect of $g$ before $k_{\text{ren}}$ is shifted and self-blocking dominates the re-
response. For a phonon at $\omega_0 = 2.2J$ [Fig. S5(d)], $u_{k_{\text{res}},0}$ is enhanced and shifted massively at strong $g$, where the downward band shift (corresponding to an upward $k_{\text{res}}$ shift) is significant and self-blocking is weak. However, it is clear from these panels that a quantitative characterization of the band renormalization caused by driving phonons at frequency $\omega_0$ using a pump at frequency $\omega$ requires the introduction of a further frequency, and for this we introduce the probe beam at $\Omega$.

In Figs. S5(e) and S5(f) we illustrate the accurate extraction of the new band edge from the probe response. In the absence of driving [Fig. S5(e)], the response in the probe spectrum peaks not at the undriven band edge but at a slightly shifted frequency. This shift, $\delta \omega_k$, is the result of weak hybridization between the triplon pairs near the band edge and the phonon at $\omega_0 = 2.2J$, and its value remains constant as the amplitude of the probe ($E_0$) is reduced to zero. When the driving electric field ($E_0$)
is restored, in Fig. [S5f] we observe a large shift in the response peaks and we deduce the true location of the renormalized band edge by subtracting $\delta \omega_s$.

In Fig. [S5b] we show the full cosine band of the triplons to illustrate its driven renormalization for different $g$ values. The phononic driving takes place at $\omega = \omega_0 = 2.2J$ and the quantity $2\tilde{\omega}_k$ gives the lower and upper band edges deduced from the pump-probe spectra of Figs. 3(b) and 3(c) of the main text when the driving electric field is doubled from our standard driving, i.e. the band edges deduced from the dotted lines in these figures. In the inset we compare the positions of the lower band edges for each value of $g$, which include the relevant $\delta \omega_s$ corrections, with the values expected from a renormalization of $J$ in Eq. (S2) to $\tilde{J} = J + g\tilde{n}_0(g)$, where $\tilde{n}_0(g)$ is taken from Fig. 3(a) of the main text. The excellent agreement indicates that any higher-order contributions to the driven band renormalization remain small by comparison with the effect of the shift in $\tilde{n}_0$ for the model we consider [Fig. 1(a) of the main text].

For further insight into the meaning of our pump-probe results, in Fig. [S5g] we show the phonon spectrum matching the spin response of Fig. [S5f], i.e. for probe frequencies around the lower band edge, and in Fig. [S5h] the phononic response for probing around the upper band edge. The strongest probe fields in these two panels match the spin spectra at $g = 0.5J$ shown in Figs. 3(b) and 3(c) of the main text. In the absence of a probe beam, the phonon spectrum is essentially flat around the band edges, with no discernible features forming in these regions when the only driving is resonant with the available in-band phonon at $\omega_0 = 2.2J$. However, increasing the probe intensity reveals that the formation of the predominantly magnetic spectral features at the band edge in Figs. 3(b), 3(c), and [S5f] is accompanied by a small dip in $n_{\text{ph}}(\omega_0, \Omega)$. This weak response indicates the weak phononic character of these hybrid states; the fact that it is negative is again a consequence of self-blocking, in that the resonant hybrid interferes weakly with the uptake of laser energy by the phonon.

S5. $J'$ MODEL

To gain further perspective on the nature of magnetophononic self-blocking and triplon band engineering, we consider a model in which the Einstein phonon is coupled to the (interdimer) $J'$ bond in Fig. 1(a) of the main text, i.e. the spin-phonon part of the Hamiltonian in Eq. (S3) is changed to

$$H_{\text{sp}} = \sum_j g(b_j + b_j^\dagger) \tilde{S}_{2,j} \cdot \tilde{S}_{1,j+1}. \quad (S14)$$

This “$J'$ model” has two primary differences from the “$J$ model” considered hitherto. First, because the spin-phonon coupling has dimensions of $J$ but the interdimer bond has magnitude $J'$, the effect of the coupling term [Eq. (S14)] is amplified in the equations of motion and it is convenient to compare the $J'$-model results to a $J$-model with the rescaled coupling $g' = gJ'/J$. Second, the coefficients in Eqs. (S9a) and (S9b) undergo the alterations $y_k \to -y_k'/\lambda$ and $y_k \to -y_k/\lambda$, which lead to specific changes in the physics.

First for self-blocking, if $g$ is replaced by $g' (= g/2$ for the illustrative parameters we use) then the results for $n_{\text{ph}}$ are numerically very close to those of Fig. 1(b) of the main text. This reinforces our statement that self-blocking is indeed a generic phenomenon in a driven spin-phonon-coupled system, rather than possibly being a special consequence of localization, dimerization, and unit-cell selection. Turning to hybrid-state formation, the phenomenology of Figs. 2 and [S4] is also reproduced in the $J'$ model, with frequency shifts that are approximately four times as large if $g$ is not replaced by $g'$. Finally, triplon band engineering in the $J'$ model is quite different because of the altered coefficients mentioned above: $\tilde{n}_0$ is smaller by one order of magnitude than the values found in Fig. 3(a) of the main text, meaning that the band-shifts of Figs. 3(b) and 3(c) are not attainable. However, $\tilde{n}_0$ also changes sign as the driving phonon frequency passes through the center of the two-triplon band ($\omega_0 = 2J$), indicating that phonons coupled to $J'$ offer a different type of band control, in the form of a band-narrowing or -broadening. These results are not surprising if one considers the expression for $\omega_s$ in Eq. (S2). Figure [S5i] shows the probe response at the lower band edge of an undriven model with $g' = 0.5J'$, from which we observe that the frequency shift, $\delta \omega_s$, is almost identical to that in Fig. [S5f]. In Fig. [S5j] we observe that the (upward) shift of the lower band edge caused by driving this model at $\omega_0 = 2.2J$ is very weak, and in fact it corresponds to a band-narrowing (the upper edge is pulled downwards). We comment for completeness that driving a phonon at $\omega = \omega_0 = 1.8J$ gives the strongest response in the $J'$ model, a band-broadening that is approximately twice as large for the same driving strength.