Possible Many-Body Localization in a Long-Lived Finite-Temperature Ultracold Quasineutral Molecular Plasma

John Sous[1,2] and Edward Grant[1,3,*]

[1]Department of Physics & Astronomy, University of British Columbia, Vancouver, British Columbia V6T 1Z3, Canada
[2]Stewart Blusson Quantum Matter Institute, University of British Columbia, Vancouver, British Columbia V6T 1Z4, Canada
[3]Department of Chemistry, University of British Columbia, Vancouver, British Columbia V6T 1Z3, Canada

(Received 15 August 2017; revised manuscript received 26 January 2018; published 14 March 2018)

We argue that the quenched ultracold plasma presents an experimental platform for studying the quantum many-body physics of disordered systems in the long-time and finite energy-density limits. We consider an experiment that quenches a plasma of nitric oxide to an ultracold system of Rydberg molecules, ions, and electrons that exhibits a long-lived state of arrested relaxation. The qualitative features of this state fail to conform with classical models. Here, we develop a microscopic quantum description for the arrested phase based on an effective many-body spin Hamiltonian that includes both dipole-dipole and van der Waals interactions. This effective model appears to offer a way to envision the essential quantum disordered nonequilibrium physics of this system.

DOI: 10.1103/PhysRevLett.120.110601

Introduction.—Quantum mechanics serves well to describe the discrete low-energy dynamics of isolated microscopic many-body systems [1]. The macroscopic world conforms with the laws of Newtonian mechanics [2]. Quantum statistical mechanics [3] bridges these realms by treating the quantum mechanical properties of an ensemble of particles statistically and characterizing the properties of the system in terms of state properties (temperature, chemical potential, etc.), in an approach that implies a complex phase space of trajectories with ergodic dynamics [4]. However, this is not always the case, and the macroscopic description of quantum many-body systems that fail to behave as expected statistically remains today as a key unsolved problem [5,6].

Ergodicity, when present in an isolated quantum many-body system, emerges as the system thermalizes in a unitary evolution that spreads information among all the subspaces of the system. The subspaces act as thermal reservoirs for each other. Most known many-body systems thermalize in this fashion, obeying the eigenstate thermalization hypothesis [4,6–10], which holds that the eigenstates of corresponding many-body Hamiltonians are thermal.

Exceptions include fine-tuned integrable systems [11], and the class of so-called many-body localized systems [6,12], which have attracted intense interest in recent years. Such systems do not thermalize at finite energy densities and are therefore nonergodic. Disorder in a landscape of interactions preserves memory of the initial local conditions for infinitely long times. Many-body localized phases cannot be understood in terms of conventional quantum statistical mechanics [13,14].

Many-body localization has been observed in deliberately engineered experimental systems with ultracold atoms in one- and two-dimensional optical lattices [15–20]. In such cases, tuning of the lattice parameters allows investigation of the phase diagram of the system as a function of disorder strength. However, such ultracold systems suffer from decoherence, confining localization to short time scales and low energy densities.

It is important to determine experimentally whether conditions exist under which many-body localization can persist for long times at finite temperatures, and to understand if such a robust macroscopic quantum many-body state can occur naturally in an interacting quantum system without deliberate tuning of experimental parameters. Such a realization could pave the way to exotic quantum effects, such as entangled macroscopic objects and localization-protected quantum order [21,22], which could have societal and technological implications [23].

Motivated by these questions, we have explored the quenched ultracold molecular plasma as an arena in which to study quantum many-body effects in the long-time and finite energy-density limits [24,25]. The ultracold plasma system offers complexity, as encountered in quantum materials, but evolves from state-selected initial conditions that allow for a description in terms of a specific set of atomic and molecular degrees of freedom.

Experimental work has recently established laboratory conditions under which a high-density molecular ultracold plasma evolves from a cold Rydberg gas of nitric oxide, adiabatically sequesters energy in a reservoir of global mass transport, and relaxes to form a spatially correlated, strongly coupled plasma [25,26]. This system naturally evolves to form an arrested phase that has a long lifetime with respect to recombination and neutral dissociation, and a very slow rate of free expansion. These volumes exhibit

state properties that are independent of the initial quantum state and density, parameters that critically affect the time scale of relaxation, suggesting a robust process of self-assembly that reaches an arrested state, far from conventional thermal equilibrium.

Departure from classical models suggests localization in the disposition of energy [25]. In an effort to explain this state of arrested relaxation, we have developed a quantum mechanical description of the system in terms of a power law interacting spin model, which allows for the possibility of slow dynamics or many-body localization.

Experiment.—The double-resonant pulsed-laser excitation of nitric oxide entrained in a supersonic molecular beam forms a characteristic Gaussian ellipsoid volume of state-selected Rydberg gas that propagates in z with a well-defined velocity, longitudinal temperature ($T_{||} = 500$ mK), transverse temperature ($T_{\perp} < 5$ mK), and precisely known initial density in a range from $\rho_0 = 10^{10}$ to 10^{12} cm^{-3} (see Fig. 1 and Refs. [27,28]).

Rydberg molecules in the leading edge of the nearest-neighbor distance distribution interact to produce NO^+ ions and free electrons [49]. Electron–Rydberg molecule collisions trigger an ionization avalanche on a time scale from nanoseconds to microseconds depending on initial density and principal quantum number n_0.

Inelastic collisions heat electrons and the system proceeds to a quasiequilibrium of ions, electrons, and high-Rydberg molecules of nitric oxide. This relaxation and the transient state it produces entirely parallels the many observations of ultracold plasma evolution in atomic systems under the conditions of a magneto-optical trap [50].

We see this avalanche unfold directly in sequences of density-classified selective field ionization spectra measured as a function of delay after the initial formation of the Rydberg gas [25]. For a moderate $\rho_0 = 3 \times 10^{11}$ cm^{-3}, the ramp-field signal of the selected Rydberg state, n_0 gives way on a 100 ns time scale to form the selective field ionization spectrum of a system in which electrons bind very weakly to single ions in a narrow distribution of high Rydberg states or in a quasifree state held by the plasma space charge [28].

The peak density of the plasma decays for as much as 10 μs until it reaches a value of $\sim 4 \times 10^{10}$ cm^{-3}, independent of the initially selected n_0 and ρ_0. Thereafter, the number of charged particles remains constant for at least a millisecond. On this hydrodynamic time scale, the plasma bifurcates, disposing substantial energy in the relative velocity of plasma volumes separating in $\pm x$, the cross-beam axis of laser propagation [26].

The avalanche to plasma proceeds at a rate predicted with accuracy by semiclassical coupled rate equations [25,28]. This picture also calls for the rapid collisional relaxation of Rydberg molecules, accompanied by an increase in the electron temperature to 60 K or more. Bifurcation accounts for a loss of electron energy. But the volumes that remain cease to evolve, quenching instead to form an arrested phase that expands slowly, at a rate reflecting an initial electron temperature no higher than a few degrees kelvin. These volumes show no sign of loss owing to the fast dissociative recombination of NO^+ ions with electrons predicted classically for low T_e [51], or predissociation of NO Rydberg molecules, well known to occur with relaxation in n [52].

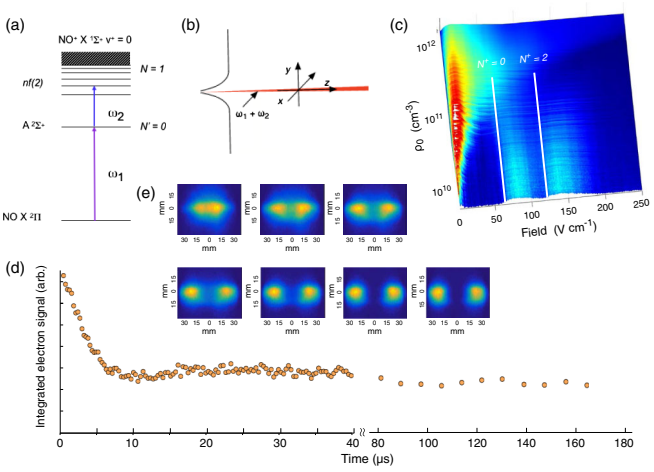

FIG. 1. (a) Double-resonant selection of the initial quantum state of the $n_0 f(2)$ Rydberg gas. (b) Laser-crossed differentially pumped supersonic molecular beam. (c) Selective field ionization spectrum after a 500 ns evolution, showing the signal of weakly bound electrons combined with a residual population of $49f(2)$ Rydberg molecules. After 10 μs, this population sharpens to signal only high-n Rydberg molecules and plasma electrons. (d) Integrated electron signal as a function of evolution time from 0 to 160 μs. Note the onset of the arrested phase before 10 μs. Time scale compressed by a factor of 2 after 80 μs. (e) x, y-integrated images recorded after a flight time of 400 μs with $n_0 = 40$ for initial Rydberg gas peak densities varying from 2×10^{11} to 1×10^{12} cm^{-3}. All of these images exhibit the same peak density: 1×10^7 cm^{-3}.

Thus, from the experiment, we learn that 5 μs after avalanche begins, Rydberg relaxation ceases. We detect no sign of ion acceleration by hot electrons and the surviving number of ions and electrons remains constant for the entire remaining observation period, extending to as long as 1 ms. With the vast phase space available to energized electrons and neutral nitrogen and oxygen atom fragments, this persistent localization of energy in the electrostatic separation of cold ions and electrons represents a very significant departure from a thermalized phase. Current experimental evidence thus points strongly to energy localization and the absence of thermalization within the accessible time of the experiment.

Molecular physics of the arrested phase.—Direct measurements of its electron binding energy together with its observed expansion rate establish experimentally that the bifurcated plasma contains only high-Rydberg molecules ($n > 80$) and NO^+ ions in combination with cold electrons (initial $T_e < 5$ K) bound by the space charge. As noted

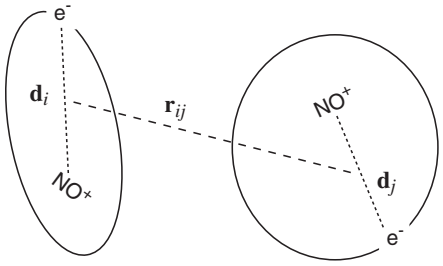

FIG. 2. Schematic representation of NO^+ core ions, paired with extravalent electrons to form interacting dipoles $\mathbf{d}_i$ and $\mathbf{d}_j$, separated by $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$.

above, semiclassical models mixing these species in any proportion predict thermal relaxation, electron heating, expansion, and dissipation on a rapid time scale with very evident consequences completely unobserved in the experiment. Instead, beyond an evolution time of 10 μs or less, we find that the plasma settles in a state of arrested relaxation of canonical density and low internal energy manifested by a slow free expansion.

To describe this apparent state of suppressed relaxation, we proceed now to develop a formal representation of the predominant interactions in this arrested phase. Under the evidently cold, quasineutral conditions of the relaxed plasma, ions pair with extravalent electrons to form dipoles that interact as represented schematically in Fig. 2.

Assuming an intermolecular spacing that exceeds the dimensions of individual ion-electron separations, we can describe the Coulomb interactions represented in Fig. 2 in terms of a simple Hamiltonian:

$$H = \sum_i \left(\frac{\mathbf{P}_i^2}{2m} + h_i \right) + \sum_{i,j} V_{ij}, \tag{1}$$

where h_i describes the local relationship of each electron with its proximal NO^+ core. This local representation extends to account for the interactions of a bound extravalent electron with vibrational, rotational, and electronic degrees of freedom of the core, as described, for example, by multichannel quantum defect theory [53]. Each ion-electron pair has momentum $\mathbf{P}_i$ and $V_{ij} \equiv V(\mathbf{r_i} - \mathbf{r_j})$ describes the potential energy of the interacting multipoles, represented in Fig. 2 to lowest order as induced dipoles with an interaction defined by $V_{ij}^{\mathrm{dd}} = [\mathbf{d}_i \cdot \mathbf{d}_j - 3(\mathbf{d}_i \cdot \mathbf{r}_{ij})(\mathbf{d}_j \cdot \mathbf{r}_{ij})]/\mathbf{r}_{ij}^3$, where for simplicity we average over the anisotropy of the dipole-dipole interaction.

The plasma also very likely includes ion-electron pairs of positive total energy. This implies the existence of local Hamiltonians of much greater complexity that define quasi-Rydberg bound states with dipole and higher-order moments formed by the interaction of an extravalent electron with more than one ion.

Representing the eigenstates of h_i by $|e_i\rangle$, we can write a reduced Hamiltonian for the pairwise dipole-dipole interactions [54,55] in the arrested phase:

$$H^{\mathrm{dd}} = \sum_i \frac{\mathbf{P}_i^2}{2m} + \sum_{i,j} V_{ij}^{\mathrm{dd}}, \tag{2}$$

where we evaluate V_{ij}^{dd} in the $|e_i\rangle$ basis.

Note that such a Hamiltonian usually refers to the case where a narrow bandwidth laser prepares a Rydberg gas in which a particular set of dipole-dipole interactions gives rise to a small, specific set of coupled states [56–58]. By contrast, the molecular ultracold plasma forms spontaneously by the processes of avalanche and quench to populate a great many different states that evolve spatially without the requirement of light-matter coherence or reference to a dipole blockade of any kind.

This system relaxes to a quenched regime of ultracold temperature, from which it expands radially at a rate of a few meters per second. Dipolar energy interactions proceed on a much faster time scale [59–62]. Cross sections for close-coupled collisions are minuscule by comparison [63]. We can thus assume that the coupled states defined by dipole-dipole interactions evolve adiabatically with the motion of ion centers.

This separation of time scales enables us to write an effective Hamiltonian describing pairwise interactions that slowly evolve in an instantaneous frame of slowly moving ions and Rydberg molecules: $H_{\mathrm{eff}} = \mathcal{P}\sum_{i,j} V_{ij}^{\mathrm{dd}}$, where $\mathcal{P}$ represents a projector onto the low-energy degrees of freedom owing to dipole-dipole coupling.

Effective many-body Hamiltonian.—Considering pairwise dipolar interactions between ion-electron pairs, we choose a set of basis states $|e^1\rangle, |e^2\rangle, \ldots, |e^L\rangle$ that spans the low-energy regime. The superscript with the lower (higher) integer label refers to the state with larger (smaller) electron binding energy.

Quenching gives rise to a vast distribution of rare resonant pairwise interactions, creating a random potential landscape. Dipole-dipole interactions in this dense manifold of basis states cause excitation exchange. In the disorder potential, these processes are dominated by low energy-excitations involving L states in number, where we expect L to be small (from 2 to 4). The most probable interactions select L-level systems composed of different basis states from dipole to dipole.

In a limit of dipole-dipole coupling, we can represent pairwise excitations by spins with energies ϵ_i and exchange interactions governed by an XY model Hamiltonian [28,64] that describes these interactions in terms of their effective spin dynamics:

$$H_{\mathrm{eff}} = \sum_i \epsilon_i \hat{S}_i^z + \sum_{i,j} J_{ij}(\hat{S}_i^+ \hat{S}_j^- + \mathrm{H.c.}), \tag{3}$$

where $\hat{S}$ in each case denotes a spin-L operator defined as $\hat{S}^{\gamma} = \hbar\hat{\sigma}^{\gamma}/2$, for which σ^{γ} is the corresponding spin-L Pauli matrix that spans the space of the L active levels and $\gamma = x$, y, or z. H.c. refers to the Hermitian conjugate.

This Hamiltonian reflects both the diagonal and off-diagonal disorder created by the variation in L-level system from dipole to dipole. The first term in H_{eff} describes the diagonal disorder arising from random contributions to the on-site energy of any particular dipole owing to its random local environment. In spin language, $\sum_i \epsilon_i \hat{S}_i^z$ represents a Gaussian-distributed random local field of width W. The representative selective field ionization spectrum in Fig. 1 directly gauges a W of ~500 GHz for the quenched ultracold plasma.

In the second term, $J_{ij} = t_{ij}/r_{ij}^3$ determines the off-diagonal disordered amplitudes of the spin flip-flops. To visualize the associated disorder, recognize that the second term varies as $t_{ij} \propto |\mathbf{d}_i||\mathbf{d}_j|$, where every interaction selects a different $\mathbf{d}_i$ and $\mathbf{d}_j$. Over the present range of W, a simple pair of dipoles formed by s and p Rydberg states of the same n couple with a t_{ij} of 75 GHz μm^3 [65]. Note that t_{ij} falls exponentially with the difference in principal quantum numbers, Δn_{ij} [66].

Induced Ising interactions.—In the limit $|J_{ij}| \ll W$ most appropriate to the experiment, sequences of interactions can add an Ising term that describes a van der Waals shift of pairs of dipoles [28,67]. These processes occur with an amplitude $U_{ij} \approx J_{ij}^2 \tilde{J}/W^2$, where $\tilde{J}$ estimates J_{ij}, for an average value of t_{ij} at an average distance separating the spins. U_{ij} is inherently random owing to the randomness in J_{ij}.

Together, these results lead us to a general spin model with dipole-dipole and van der Waals interactions [28]:

$$H_{\text{eff}} = \sum_i \epsilon_i \hat{S}_i^z + \sum_{i,j} J_{ij}(\hat{S}_i^+ \hat{S}_j^- + \text{H.c.}) + \sum_{i,j} U_{ij} \hat{S}_i^z \hat{S}_j^z, \tag{4}$$

where $U_{ij} = D_{ij}/r_{ij}^6$ and $D_{ij} = t_{ij}^2 \tilde{J}/W^2$.

Discussion: localization versus glassy behavior and slow dynamics.—The complexity of this Hamiltonian places an exact solution of Eq. (4) beyond reach for the conditions of the plasma. But we can gauge some likely properties of such a solution by analogy to published work on simpler systems.

In the single-spin limit, this Hamiltonian reduces to the dipolar XY model, which has been studied by locator expansion methods measuring the probability of resonant pairs [68,69]. When J_{ij} scales by a power law α that equals the dimension d, a single-spin model with diagonal disorder displays critical behavior characterized by extended states with subdiffusive dynamics [68,69]. Dipolar systems in three dimensions can form extended states, but yet exhibit nonergodic behavior [70].

Off-diagonal disorder in the presence of long-range spin flip-flop interactions of arbitrary order in one dimension yields algebraic localization as opposed to exponential Anderson localization, challenging the generality of the rule that says systems must delocalize for $\alpha \leq d$ [71].

The many-body problem is more involved, because the van der Waals term forms off-diagonal matrix elements in the resonant pair states [72]. This mechanism couples distant resonant pairs, transferring energy from one pair to the other to cause delocalization. A study of power-law coupled systems predicts that spin flip-flops (order α) and spin Ising interactions (order β) in an iterated pair configuration in which $\beta \leq \alpha$ will localize for $\beta/2 > d$ [73].

A locator expansion approach developed for $\beta > \alpha$ applied to Eq. (4) confined to diagonal disorder predicts a critical dimension $d_c = 2$ [67]. For the case of $d > d_c$, this theory holds that a diverging number of resonances drives delocalization whenever the number of dipoles exceeds a critical value N_c.

A system described by Eq. (4) for the conditions under which we observe arrest requires a number of dipoles $N_c = (W/\tilde{J})^4 \approx 3 \times 10^9$ to delocalize [28]. This theoretical threshold deemed necessary for resonance delocalization exceeds the number of molecules found experimentally in the quenched ultracold plasma by more than an order of magnitude [28].

Moreover, as Nandkishore and Sondhi have pointed out [74], locator expansion arguments might not hold generally, and low-order power law interactions could well give rise to many-body localization in higher dimensions. Their arguments build on the idea that, in many systems, long-range interactions can drive a system to form correlated phases in which emergent short-range interactions can be well characterized by a locator expansion perturbation theory approach. In this context, many-body localization with long-range interactions in higher dimensions becomes quite possible.

A related study has investigated the behavior of a three-dimensional dipolar system of nitrogen-vacancy color centers in diamond in the presence of on-site disorder [75]. The experimental results point to slow dynamics consistent with our observations.

The forgoing analysis suggests that the model defined by Eq. (4) ought to exhibit some form of localization or at least very slow dynamics, since all the terms in the Hamiltonian are disordered and the terms responsible for delocalization (J_{ij} and U_{ij}) are expected to be much smaller than W. This seems to be what we see in the experiment.

Concluding remarks.—This work has argued that the quenched ultracold plasma forms an arrested phase possibly governed by quantum disordered nonequilibrium physics in long-time and finite energy-density limits. In an effort to support this notion, we have suggested that the evident and certainly present quantum dipolar interactions can be usefully described by a disordered spin model and

have analyzed its properties in the limit of strong on-site disorder by analogy with theoretical results for simpler dipolar systems.

Considering the challenge of scale confronting the accurate numerical solution of large disordered problems and the apparent contradiction of available theoretical results [76–79], experimental systems stand to play an important role in understanding localization and slow dynamics. The results presented here call in particular for further experimental and theoretical efforts to probe the physics of localization in long-range interacting systems of higher dimension. The quenched ultracold plasma appears to offer a view of large-scale quantum dynamics in a regime inaccessible to optical lattices and solid-state materials.

This work was supported by the U.S. Air Force Office of Scientific Research (Grant No. FA9550-17-1-0343), together with the Natural Sciences and Engineering research Council of Canada (NSERC) and the Stewart Blusson Quantum Matter Institute (SBQMI). J. S. gratefully acknowledges support from the Harvard-Smithsonian Institute for Theoretical Atomic, Molecular and Optical Physics (ITAMP). We have benefited from helpful interactions with Rahul Nandkishore, Shivaji Sondhi, and Alexander Burin. We also appreciate discussions with Joshua Cantin and Roman Krems.

*To whom all correspondence (inquiry) should be addressed. edgrant@chem.ubc.ca

[1] J. J. Sakurai, *Modern Quantum Mechanics* (Pearson, London, 2014).
[2] H. Goldstein, *Classical Mechanics* (Pearson Education, India, 2011).
[3] M. Kardar, *Statistical Physics of Particles* (Cambridge University Press, Cambridge, England, 2007).
[4] L. D'Alessio, Y. Kafri, A. Polkovnikov, and M. Rigol, From quantum chaos and eigenstate thermalization to statistical mechanics and thermodynamics, Adv. Phys. **65**, 239 (2016).
[5] A. Polkovnikov, K. Sengupta, A. Silva, and M. Vengalattore, Colloquium: Nonequilibriumdynamics of closed interacting quantum systems, Rev. Mod. Phys. **83**, 863 (2011).
[6] R. Nandkishore and D. A. Huse, Many-body localization and thermalization in quantum statistical mechanics, Annu. Rev. Condens. Matter Phys. **6**, 15 (2015).
[7] J. M. Deutsch, Quantum statistical mechanics in a closed system, Phys. Rev. A **43**, 2046 (1991).
[8] H. Tasaki, From Quantum Dynamics to the Canonical Distribution: General Picture and a Rigorous Example, Phys. Rev. Lett. **80**, 1373 (1998).
[9] M. Srednicki, Chaos and quantum thermalization, Phys. Rev. E **50**, 888 (1994).
[10] M. Rigol, V. Dunjko, and M. Olshanii, Thermalization and its mechanism for generic isolated quantum systems, Nature (London) **452**, 854 (2008).
[11] M. Rigol, Breakdown of Thermalization in Finite One-Dimensional Systems, Phys. Rev. Lett. **103**, 100403 (2009).
[12] D. Basko, I. Aleiner, and B. Altshuler, Metal—insulator transition in a weakly interacting many-electron system with localized single-particle states, Ann. Phys. (Amsterdam) **321**, 1126 (2006).
[13] R. Vosk, D. A. Huse, and E. Altman, Theory of the Many-Body Localization Transition in One-Dimensional Systems, Phys. Rev. X **5**, 031032 (2015).
[14] V. Khemani, S. P. Lim, D. N. Sheng, and D. A. Huse, Critical Properties of the Many-Body Localization Transition, Phys. Rev. X **7**, 021013 (2017).
[15] S. S. Kondov, W. R. McGehee, W. Xu, and B. DeMarco, Disorder-Induced Localization in a Strongly Correlated Atomic Hubbard Gas, Phys. Rev. Lett. **114**, 083002 (2015).
[16] M. Schreiber, S. S. Hodgman, P. Bordia, H. P. Lüschen, M. H. Fischer, R. Vosk, E. Altman, U. Schneider, and I. Bloch, Observation of many-body localization of interacting fermions in a quasi-random optical lattice, Science **349**, 842 (2015).
[17] J. Smith, A. Lee, P. Richerme, B. Neyenhuis, P. W. Hess, P. Hauke, M. Heyl, D. A. Huse, and C. Monroe, Many-body localization in a quantum simulator with programmable random disorder, Nat. Phys. **12**, 907 (2016).
[18] J.-Y. Choi, S. Hild, J. Zeiher, P. Schauß, A. Rubio-Abadal, T. Yefsah, V. Khemani, D. A. Huse, I. Bloch, and C. Gross, Exploring the many-body localization transition in two dimensions, Science **352**, 1547 (2016).
[19] H. P. Lüschen, P. Bordia, S. S. Hodgman, M. Schreiber, S. Sarkar, A. J. Daley, M. H. Fischer, E. Altman, I. Bloch, and U. Schneider, Signatures of Many-Body Localization in a Controlled Open Quantum System, Phys. Rev. X **7**, 011034 (2017).
[20] P. Bordia, H. Lüschen, S. Scherg, S. Gopalakrishnan, M. Knap, U. Schneider, and I. Bloch, Probing Slow Relaxation and Many-Body Localization in Two-Dimensional Quasiperiodic Systems, Phys. Rev. X **7**, 041047 (2017).
[21] D. A. Huse, R. Nandkishore, V. Oganesyan, A. Pal, and S. L. Sondhi, Localization-protected quantum order, Phys. Rev. B **88**, 014206 (2013).
[22] R. Vasseur, S. A. Parameswaran, and J. E. Moore, Quantum revivals and many-body localization, Phys. Rev. B **91**, 140202 (2015).
[23] N. Y. Halpern, C. D. White, S. Gopalakrishnan, and G. Refael, Mbl-mobile: Many-body-localized engine, arXiv: 1707.07008.
[24] J. P. Morrison, C. J. Rennick, J. S. Keller, and E. R. Grant, Evolution from a Molecular Rydberg Gas to an Ultracold Plasma in a Seeded Supersonic Expansion of NO, Phys. Rev. Lett. **101**, 205005 (2008).
[25] R. Haenel, M. Schulz-Weiling, J. Sous, H. Sadeghi, M. Aghigh, L. Melo, J. S. Keller, and E. R. Grant, Arrested relaxation in an isolated molecular ultracold plasma, Phys. Rev. A **96**, 023613 (2017).
[26] M. Schulz-Weiling and E. R. Grant, Quantum state control of ultracold plasma fission, J. Phys. B **49**, 064009 (2016).
[27] M. Schulz-Weiling, H. Sadeghi, J. Hung, and E. R. Grant, On the evolution of the phase-space distributions of a non-spherical molecular ultracold plasma in supersonic beam, J. Phys. B **49**, 193001 (2016).

[28] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.110601 for details, which includes Refs. [29–48].
[29] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, Cambridge, England, 2005).
[30] R. Patel, N. Jones, and H. Fielding, Rotational-state-selective field ionization of molecular Rydberg states, Phys. Rev. A **76**, 043413 (2007).
[31] P. Mansbach and J. Keck, Monte carlo trajectory calculations of atomic excitation and ionization by thermal electrons, Phys. Rev. **181**, 275 (1969).
[32] T. Pohl, D. Vrinceanu, and H. R. Sadeghpour, Rydberg Atom Formation in Ultracold Plasmas: Small Energy Transfer with Large Consequences, Phys. Rev. Lett. **100**, 223201 (2008).
[33] M. Bixon and J. Jortner, Intramolecular coupling between non-penetrating high molecular Rydberg states, Molecular Physics **89**, 373 (1996).
[34] E. Murgu, J. D. D. Martin, and T. F. Gallagher, Stabilization of predissociating nitric oxide Rydberg molecules using microwave and radio-frequency fields, J. Chem. Phys. **115**, 7032 (2001).
[35] W. A. Chupka, Factors affecting lifetimes and resolution of Rydberg states observed in zero-electron-kinetic-energy spectroscopy, J. Chem. Phys. **98**, 4520 (1993).
[36] N. Saquet, J. P. Morrison, M. Schulz-Weiling, H. Sadeghi, J. Yiu, C. J. Rennick, and E. R. Grant, On the formation and decay of a molecular ultracold plasma, J. Phys. B **44**, 184015 (2011).
[37] N. Saquet, J. P. Morrison, and E. Grant, Recombinative dissociation and the evolution of a molecular ultracold plasma, J. Phys. B **45**, 175302 (2012).
[38] D. S. Dorozhkina and V. E. Semenov, , Exact Solution of Vlasov Equations for Quasineutral Expansion of Plasma Bunch into Vacuum, Phys. Rev. Lett. **81**, 2691 (1998).
[39] H. Sadeghi and E. R. Grant, Dissociative recombination slows the expansion of a molecular ultracold plasma, Phys. Rev. A **86**, 052701 (2012).
[40] V. M. Agranovich, *Excitations in Organic Solids*, Vol. 142 (Oxford University Press, New York, 2009).
[41] J. M. Brown and A. Carrington, *Rotational Spectroscopy of Diatomic Molecules* (Cambridge University Press, Cambridge, England, 2003).
[42] J. H. Gurian, P. Cheinet, P. Huillery, A. Fioretti, J. Zhao, P. L. Gould, D. Comparat, and P. Pillet, Observation of a Resonant Four-Body Interaction in Cold Cesium Rydberg Atoms, Phys. Rev. Lett. **108**, 023005 (2012).
[43] H. Zoubi, A. Eisfeld, and S. Wüster, Van der Waals–stabilized Rydberg aggregates, Phys. Rev. A **89**, 053426 (2014).
[44] J. Z. Imbrie, Diagonalization and Many-Body Localization for a Disordered Quantum Spin Chain, Phys. Rev. Lett. **117**, 027201 (2016).
[45] S. Gopalakrishnan, M. Müller, V. Khemani, M. Knap, E. Demler, and D. A. Huse, Low-frequency conductivity in many-body localized systems, Phys. Rev. B **92**, 104202 (2015).
[46] S. Gopalakrishnan, K. Agarwal, E. A. Demler, D. A. Huse, and M. Knap, Griffiths effects and slow dynamics in nearly many-body localized systems, Phys. Rev. B **93**, 134206 (2016).
[47] K. Agarwal, E. Altman, E. Demler, S. Gopalakrishnan, D. A. Huse, and M. Knap, , Rare-region effects and dynamics near the many-body localization transition, Ann. Phys. (Amsterdam) **529**, 1600326 (2017).
[48] P. Ponte, C. R. Laumann, D. A. Huse, and A. Chandran, Thermal inclusions: how one spin can destroy a many-body localized phase, Phil. Trans. R. Soc. A **375**, 20160428 (2017).
[49] H. Sadeghi, A. Kruyen, J. Hung, J. H. Gurian, J. P. Morrison, M. Schulz-Weiling, N. Saquet, C. J. Rennick, and E. R. Grant, Dissociation and the Development of Spatial Correlation in a Molecular Ultracold Plasma, Phys. Rev. Lett. **112**, 075001 (2014).
[50] A. Walz-Flannigan, J. R. Guest, J. H. Choi, and G. Raithel, Cold-Rydberg-gas dynamics, Phys. Rev. A **69**, 063405 (2004).
[51] I. F. Schneider, I. Rabadán, L. Carata, L. Andersen, A. Suzor-Weiner, and J. Tennyson, Dissociative recombination of NO: calculations and comparison with experiment, J. Phys. B **33**, 4849 (2000).
[52] F. Remacle and M. Vrakking, Decay dynamics of the predissociating high Rydberg states of NO, J. Phys. Chem. A **102**, 9507 (1998).
[53] C. H. Greene and C. Jungen, Molecular applications of quantum defect theory, in *Advances in Atomic and Molecular Physics*, Vol. 21 (Elsevier, New York, 1985), pp. 51–121.
[54] M. Baranov, M. Dalmonte, G. Pupillo, and P. Zoller, Condensed matter theory of dipolar quantum gases, Chem. Rev. **112**, 5012 (2012).
[55] R. Krems, B. Friedrich, and W. C. Stwalley, *Cold Molecules: Theory, Experiment, Applications* (CRC Press, 2009).
[56] P. Pillet and D. Comparat, Dipole blockade in a cold Rydberg atomic sample [Invited], J. Opt. Soc. Am. B **27**, A208 (2010).
[57] R. Löw, H. Weimer, J. Nipper, J. B. Balewski, B. Butscher, H. P. Büchler, and T. Pfau, An experimental and theoretical guide to strongly interacting Rydberg gases, J. Phys. B **45**, 113001 (2012).
[58] O. Firstenberg, C. S. Adams, and S. Hofferberth, Nonlinear quantum optics mediated by Rydberg interactions, J. Phys. B **49**, 152003 (2016).
[59] O. Mülken, A. Blumen, T. Amthor, C. Giese, M. Reetz-Lamour, and M. Weidemüller, Survival Probabilities in Coherent Exciton Transfer with Trapping, Phys. Rev. Lett. **99**, 090601 (2007).
[60] G. Günter, H. Schempp, M. Robert-de Saint-Vincent, V. Gavryusev, S. Helmrich, C. S. Hofmann, S. Whitlock, and M. Weidemüller, Observing the dynamics of dipole-mediated energy transport by interaction-enhanced imaging, Science **342**, 954 (2013).
[61] D. Barredo, H. Labuhn, S. Ravets, T. Lahaye, A. Browaeys, and C. S. Adams, Coherent Excitation Transfer in a Spin Chain of Three Rydberg Atoms, Phys. Rev. Lett. **114**, 113002 (2015).
[62] J. Zeiher, J.-Y. Choi, A. Rubio-Abadal, T. Pohl, R. van Bijnen, I. Bloch, and C. Gross, Coherent Many-Body Spin Dynamics in a Long-Range Interacting Ising Chain, Phys. Rev. X **7**, 041063 (2017).
[63] T. Michaelsen, B. Bastian, E. Carrascosa, J. Meyer, D. H. Parker, and R. Wester, Imaging state-to-state reactive

scattering in the $Ar^+ + H_2$ charge transfer reaction, J. Chem. Phys. **147**, 013940 (2017).
[64] S. Sachdev, *Quantum Phase Transitions* (Wiley Online Library, 2007).
[65] H. Zoubi, Van der Waals interactions among alkali Rydberg atoms with excitonic states, J. Phys. B **48**, 185002 (2015).
[66] N. Samboy, Long-range interactions between rubidium and potasium Rydberg atoms, Phys. Rev. A **95**, 032702 (2017).
[67] A. L. Burin, Localization in a random xy model with long-range interactions: Intermediate case between single-particle and many-body problems, Phys. Rev. B **92**, 104428 (2015).
[68] P. W. Anderson, Absence of diffusion in certain random lattices, Phys. Rev. **109**, 1492 (1958).
[69] L. Levitov, Critical hamiltonians with long range hopping, Ann. Phys. (N.Y.) **8**, 697 (1999).
[70] X. Deng, B. L. Altshuler, G. V. Shlyapnikov, and L. Santos, Quantum Levy Flights and Multifractality of Dipolar Excitations in a Random System, Phys. Rev. Lett. **117**, 020401 (2016).
[71] X. Deng, V. Kravtsov, G. Shlyapnikov, and L. Santos, Algebraic localization in disordered one-dimensional systems with long-range hopping, arXiv:1706.04088.
[72] A. L. Burin, Energy delocalization in strongly disordered systems induced by the long-range many-body interaction, arXiv:cond-mat/0611387.
[73] N. Y. Yao, C. R. Laumann, S. Gopalakrishnan, M. Knap, M. Müller, E. A. Demler, and M. D. Lukin, Many-Body Localization in Dipolar Systems, Phys. Rev. Lett. **113**, 243002 (2014).
[74] R. M. Nandkishore and S. L. Sondhi, Many Body Localization with Long Range Interactions, Phys. Rev. X **7**, 041021 (2017).
[75] G. Kucsko, S. Choi, J. Choi, P. C. Maurer, H. Sumiya, S. Onoda, J. Isoya, F. Jelezko, E. Demler, N. Y. Yao *et al.*, Critical thermalization of a disordered dipolar spin system in diamond, arXiv:1609.08216.
[76] A. Chandran, A. Pal, C. R. Laumann, and A. Scardicchio, Many-body localization beyond eigenstates in all dimensions, Phys. Rev. B **94**, 144203 (2016).
[77] S. Inglis and L. Pollet, Accessing Many-Body Localized States Through the Generalized Gibbs Ensemble, Phys. Rev. Lett. **117**, 120402 (2016).
[78] W. De Roeck and F. Huveneers, Stability and instability towards delocalization in many-body localization systems, Phys. Rev. B **95**, 155129 (2017).
[79] V. K. Varma, A. Lerose, F. Pietracaprina, J. Goold, and A. Scardicchio, Energy diffusion in the ergodic phase of a many body localizable spin chain, J. Stat. Mech. (2017) 053101.