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

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We argue that the quenched ultracold plasma presents an experimental platform for studying 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 non-equilibrium physics of this system.

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 (ETH) [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 (MBL) systems [6, 12], which have attracted intense interest in recent years. Such systems do not thermalize at finite energy densities and are therefore non-ergodic. Disorder in a landscape of interactions preserves memory of the initial local conditions for infinitely long times. MBL phases cannot be understood in terms of conventional quantum statistical mechanics [13, 14].

MBL 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 timescales and low energy densities.

It is important to determine experimentally whether conditions exist under which MBL 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 initial quantum state and density, parameters which critically affect the timescale 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 power law interacting spin model, which allows for the possibility of slow dynamics or MBL

Experiment.— 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 Figure 1 and References [27, 28]).

Rydberg molecules in the leading edge of the nearest-neighbour distance distribution interact to produce NO$^+$ ions and free electrons [29]. Electron-Rydberg 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 quasi-equilibrium 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 (MOT) [30].

We see this avalanche unfold directly in sequences of density-classified selective field ionization spectra measured as a function of delay after 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 timescale to form the selective field ionization (SFI) spectrum of a system in which electrons bind very weakly to single ions in a narrow distribution of high Rydberg states or in a quasi-free 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 timescale, 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 semi-classical coupled rate equations [25, 28]. This picture also calls for the rapid collisional relaxation of Rydberg molecules, accompanied by an increase in 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 [31], or predissociation of NO Rydbergs, well-known to occur with relaxation in n [32].
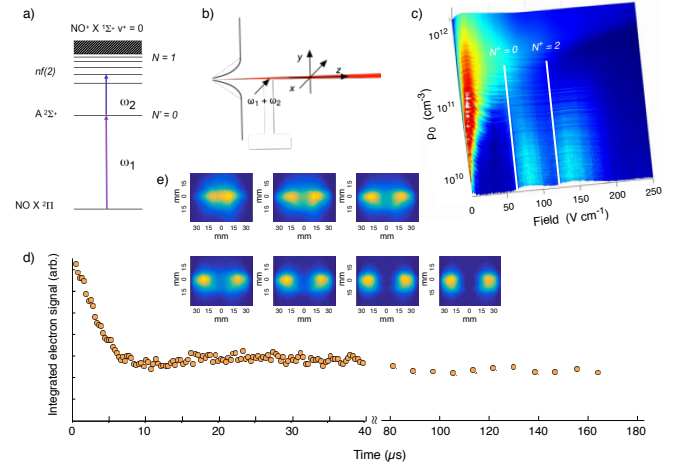

FIG. 1. a) Double-resonant selection of the initial quantum state of the $n_0 f(2)$ Rydberg gas. b) Laser-crossed differentially pumped supersonic molecular beam. c) Selective field ionization spectrum after 500 ns evolution, showing the signal of weakly bound electrons combined with a residual population of $49 f(2)$ Rydberg molecules. After 10 μs, this population sharpens to signal only high-n Rydbergs and plasma electrons. d) Integrated electron signal as a function of evolution time from 0 to 160 μs. Note the onset of the arrest phase before 10 μs. Timescale compressed by a factor of two 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 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 above, semi-classical models mixing these species in any proportion predict thermal relaxation, electron heating, expansion and dissipation on a rapid timescale with very evident consequences completely unobserved in the experiment. Instead, beyond an evolution time of 10 μs or less, we find that
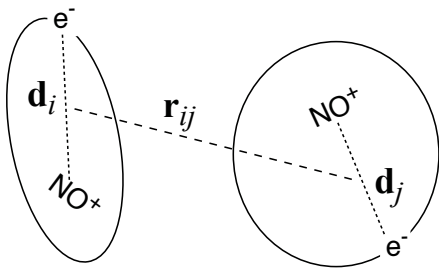
FIG. 2. Schematic representation of NO^+ core ions, paired with extravalent electrons to form interacting dipoles $\mathbf{d}_i$ and $\mathbf{d}_j$, separated by $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$.

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, quasi-neutral conditions of the relaxed plasma, ions pair with extravalent electrons to form dipoles which interact as represented schematically in Figure 2.

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

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

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 [33]. 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 Figure 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 [34, 35] in the arrested phase:

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

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 give rise to a small, specific set of coupled states [36–38]. By contrast, the molecular ultracold plasma forms spontaneously by 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 timescale [39–42]. Cross sections for close-coupled collisions are minuscule by comparison [43]. We can thus assume that the coupled states defined by dipole-dipole interactions evolve adiabatically with the motion of ion centres.

This separation of timescales enables us to write an effective Hamiltonian describing pairwise interactions that slowly evolve in an instantaneous frame of slowly moving ions and Rydberg molecules: $H_{\mathrm{eff}} = \mathcal{P} \sum_{i,j} V_{ij}^{dd}$, 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 $|e^1\rangle$, $|e^2\rangle$, ... $|e^L\rangle$ that spans the low-energy regime. The superscript with lower (higher) integer label refers to the state with larger (smaller) electron binding energy.

Quenching gives rise to a vast distribution of rare resonant pair-wise 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, 44] that describes these interactions in terms of their effective spin dynamics:

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

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 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 SFI spectrum in Figure 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 [45]. Note that t_{ij} falls exponentially with the difference in principal quantum numbers, Δn_{ij} [46].

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, 47]. These processes occur with an amplitude, $U_{ij} \approx J_{ij}^2 \widetilde{J}/W^2$, where $\widetilde{J}$ estimates J_{ij}, for an average value of t_{ij} at an average distance separating 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]:

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

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

Discussion: Localization versus glassy behaviour 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 [48, 49]. When J_{ij} scales by a power law α that equals the dimension d, a single-spin model with diagonal disorder displays critical behaviour characterized by extended states with subdiffusive dynamics [48, 49]. Dipolar systems in three dimensions can form extended states, but yet exhibit non-ergodic behaviour [50].

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 $\alpha \leq d$ [51].

The many-body problem is more involved, because the van der Waals term forms off-diagonal matrix elements in the resonant pair states [52]. 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 pairs configuration in which $\beta \leq \alpha$ will localize for $\beta/2 > d$ [53].

A locator expansion approach developed for $\beta > \alpha$ applied to Eq (4) confined to diagonal disorder predicts a critical dimension, $d_c = 2$ [47]. 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, $N_c = (W/\widetilde{J})^4 \approx 3 \times 10^9$ 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 [54], locator expansion arguments might not hold generally, and low-order power law interactions could well give rise to MBL 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, MBL 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 onsite disorder [55]. 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 non-equilibrium 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 analyzed its properties in the limit of strong onsite 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 [56–59], 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.

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