Self-induced spin glass state in elemental and crystalline neodymium

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Spin glasses are a highly complex magnetic state of matter, intricately linked to spin frustration and structural disorder. They exhibit no long-range order and exude aging phenomena, distinguishing them from quantum spin liquids. We report a new type of spin glass state, the spin-Q glass, observable in bulk-like crystalline metallic neodymium thick films. Using spin-polarized scanning tunneling microscopy combined with ab initio calculations and atomistic spin-dynamics simulations, we visualize the variations in atomic-scale non-collinear order and its response to magnetic field and temperature. We quantify the aging phenomena relating the glassiness to crystalline symmetry and the energy landscape. This result not only resolves the long-standing debate of the magnetism of neodymium, but also suggests that glassiness may arise in other magnetic solids lacking extrinsic disorder.

Spin glasses are one of the more intriguing, but least understood magnetic states of matter (*1-5*). Ferromagnets or antiferromagnets form a long-range ordered state when cooled, but spin glasses form a state characterized by seemingly random and uncorrelated magnetic patterns. The magnetization pattern in spin glasses can be compared to the amorphous structure of glasses like silicon dioxide that exhibit local structural correlations but lack a long-range ordered state. Interest in spin glasses spans many fields, ranging from iron-based superconductors (*6*) to theoretical machine learning (*4, 5, 7*), and it has been suggested to be relevant in quantum topological excitations.

Spin glasses are characterized by a glass transition temperature and by aging, that is, the magnetic state depends on its history, driven by a distribution of distinctive spin-relaxation processes with time scales spanning many orders of magnitude (*4, 5*). Aging also distinguishes spin glasses from so-called quantum spin liquids (*8*) or spin ices (*9*), which remain disordered down to zero temperature because of quantum fluctuations and lack of memory. The paradigm of all of these types of complex magnets involves magnetic frustration, derived from geometry or competing interactions. However, unlike spin liquids, disorder is traditionally also considered necessary to drive non-ergodic behavior in spin glasses.

There is still no clear understanding when spin glass behavior can arise in magnetic materials. The most commonly debated models (*1, 3-5*) describe randomly distributed spins with long-range magnetic interactions of alternating ferromagnetic and antiferromagnetic coupling, like that seen in prototypical materials such as dilute magnetic alloys (*1, 10*). In the thermodynamic limit, spin glasses have a hierarchical energy landscape with infinitely many local energy minima separated by energy barriers of multiple heights so that there is a broad distribution of transition times between different minima (*3-5, 11*) that results in an absence of local and long-range order. Most models of spin glasses invoke structural disorder, i.e. amorphousness, as a key requirement along with competing magnetic interactions (*12*). However, self-induced glassiness, a concept introduced initially for the stripe glass behavior in high-temperature superconductors (*13, 14*), was later developed for magnets (*15, 16*). Within this framework, competing interactions alone can lead to the glassy state, even in the absence of external disorder, as well as can give rise to intermediate regimes which exhibit multi-well potentials (*17*).

We show that single-crystalline elemental neodymium exhibits a new type of spin glass behavior. Spin-polarized scanning tunneling microscopy on the (0001) surface of thick Nd films revealed that the magnetic state exhibited strong local non-collinear magnetic order while lacking a long-range ordered state. This local order is defined by a spectral distribution of degenerate magnetic wave vectors, or Q states, which varied spatially and with time. We probed the response of this so-called spin-Q glass to applied magnetic fields and variable temperature, to quantify its aging behavior and energy landscape. Harnessing ab initio methods and simulations, we quantified the competing long-range magnetic interactions and the favorable Q states, illustrating that this unconventional glassy behavior results from valley-like pockets of degenerate Q states as proposed for self-induced spin glasses (*15, 16*). Moreover, we performed calculations of the autocorrelation function for pristine neodymium which also found multiple relaxation times in its spin dynamics. Our findings not only suggest that glassy behavior and aging can be found in systems with crystalline order, but also unravels an unresolved debate about the magnetic ground state of elemental Nd that has challenged scientists for several decades.

The magnetic ground state of Nd(0001)

Despite more than 50 years of investigations, there is still no consensus on the magnetic ground state of Nd and the origin of the complexity reported in various experimental observations (*18-26*). Below the Néel temperature (T_N), neutron diffraction observed the onset of static multi-Q states with decreasing temperature. Yet, it is not well understood how the multiplicity of these Q states depends on the exchange landscape of the material and which real-space magnetic interactions causes them. Moreover, other measurements indicated additional phase transitions below T_N, suggesting that the original conclusions from neutron diffraction of a modulated antiferromagnetic structure were oversimplified (*23*). In addition, there are experimental observations above T_N that give evidence for short-range order as well as a strongly frustrated exchange landscape (*26, 27*). These open questions illustrate a need for a characterization of the exchange interactions in Nd, as well as a real-space characterization of the local magnetic order.

Atomic-scale visualization of the spin-Q glass state

To characterize the atomic-scale magnetization of the surface of Nd(0001), we utilize spin-polarized scanning tunneling microscopy and spectroscopy (SP-STM/SP-STS) (*28*). We epitaxially grow thick films (up to 100 monolayers (ML)) of Nd(0001)/W(110). We note that lanthanide films prepared in this way on various bcc(110) substrates exhibit high crystallinity and superior surface cleanliness over sputter-annealed bulk single crystals (*29-37*). It has been shown for various lanthanide elements, including Nd, that films grow with bulk lattice parameters above a thickness of 10 ML (*31, 35, 38-40*), exhibit a bulk-like electronic structure above 30 ML (*32, 33, 41*), and start to exhibit bulk-like magnetic behavior in the range of 30-50 ML (*42-45*). Superlattice studies showed that 33-39 ML-thick Nd films already exhibit the temperature-dependent neutron scattering features as known from bulk single crystals (*38, 46, 47*).

The sample morphology can be tuned to either layer-by-layer grown closed films or islands (Stranski-Krastanov (SK) growth), depending on the annealing temperature (*39, 42, 48-50*). We grew two types of samples: SK grown islands of >50 ML thickness (Fig. 1A,B) and closed ~100 ML films (Fig. S1B) (*50*). As discussed above, SK islands should readily represent bulk-like structural, electronic and magnetic properties. We verified this on thicker (~100 ML) closed films, which showed identical magnetization patterns as we subsequently discuss (see supplementary text, section S2) (*50*). However, we observe that closed thick films show inferior surface qualities, due to the presence of more impurities as well as screw dislocations that can lead to pinning of magnetic structures (*51*). As the latter was not present for our island-grown samples (Fig. S1A), we focus on data taken from islands with thickness between 58 and 92 ML and lateral sizes between 58000 and 200000 nm^2, and may account for differences in previous measurements. Further details of the sample preparation and morphology as well as a discussion of thin films vs. bulk samples can be found in the supplementary text, section S1 (*50*).

Using scanning tunneling spectroscopy (STS) at low temperature (0.03−7 K), we probed the Nd(0001) surface state, whose presence and sharpness has been shown to be a probe of the cleanliness of the film (*27, 31, 32, 52, 53*). The surface state is characterized by an exchange splitting into a majority peak visible below the Fermi energy (E_F, V_S = 0) and a minority peak above E_F, with an additional narrow peak

at E_F (Fig. 1C) (*49*). We note that we saw no difference between spectra taken on the islands and on locally clean areas of the thick film. Furthermore, the magnetic exchange splitting reproduced previous low-temperature data taken on a 30 ML film (*27, 54*), which is further evidence that our samples are beyond the thin-film limit and fully reflect bulk electronic and magnetic properties.

In order to get magnetic contrast, we utilized the spin-polarized nature of the exchange-split surface state, and we imaged the surface in constant-current mode at two characteristic voltages representing dominant tunneling into the minority state (V_S = 200 mV) and out of the majority state (V_S = -150 mV), respectively (Fig. S2) (*50*). We used an out-of-plane sensitive antiferromagnetic Cr probe, which relates spin-dependent contrast variations directly to the *z*-projection, that is, the *c*-axis in the double hexagonally close packed (dhcp) structure, of the magnetization. We consider the subtracted image in order to remove stronger topographic modulations resulting from buried substrate steps or locally varying sample thickness. We refer to this subtracted image as the magnetization image (Fig. 1D,G and Fig. S2D). More details on this image processing procedure can be found in the supplementary text, section S2) (*50*).

The magnetization images of the surface revealed strong and clear short-range magnetic order with periodicities λ varying from 0.9 to 4.5 nm and oriented along or near the high-symmetry axes. These atomic-scale variations are directly related to local non-collinear magnetic order, with varying periodicities depending on spatial location, defined by a superposition of local magnetic wave vectors $Q_i = 2\pi/\lambda_i$ (Fig. 1G,H; supplementary text, section S3) (*50*). Remarkably, although clear short-range order could be seen, defined by a local multi-Q state, there was no observable long-range ordered state found for any of the probed experimental conditions.

In order to better visualize the variations of local multi-Q order, we consider reciprocal-space images obtained through Fast Fourier Transform (FFT) of the real-space magnetization images, which we refer to as Q-space images. A signature of the lack of long-range order and competing short-range order can be directly visualized by the smeared and broadly distributed spectral weight in various regions in Q space (Fig. 1E). We note that Q vectors here were derived from real-space SP-STM maps, and not directly

measured as in the case of neutron diffraction (see supplementary text, section S3 for further discussion) (*50*). We only produced and analyzed Q-space images from within a flat terrace on individual islands (typical width ≈ 150 nm). Thus, the measured broadening resulted from the spectral distribution of Q, and not from averaging over different islands with different orientations.

To highlight this, Q-space images were also produced from smaller spatial regions of the same image, which illustrated sharpened and characteristic spectral weight of Q vectors compared to the larger scale image (Figs. 1G, S4). Various line-cuts along the $\bar{\Gamma}$-$\bar{M}$ direction of multiple Q-space images taken from different spatial regions of Fig. 1D are illustrated in Fig. 1F. The resultant plots revealed a spatial variation in spectral weight in at least three distinct regions, or Q-pockets, with substantial spectral weight along the high-symmetry axes.

In the ensuing discussion, we focus particularly on the three Q-pockets with wave vectors at around Q_A = 1.1-2.0 nm^{-1}, Q_B = 2.7-3.5 nm^{-1} and Q_C = 4.6-5.3 nm^{-1}. From this information, we plot a schematic of the out-of-plane projected magnetization with respect to the atomic lattice (Fig. 1H) for the two regions shown in Fig. 1G with the defined Q vectors from these Q-pockets. The corresponding Q-space images are a direct visualization of the multi-Q nature acquired over the spatial area of the given images, and they provide a quantitative comparison to previous neutron diffraction studies (*18-21, 24-26*). The spectral weight around these pockets, as well as blurring of the intensity of the Q states in larger scale images (Fig. 1E) was an initial indication of the glassy nature of the magnetic state, and reminiscent of spin-based analogs of stripe or checkerboard order in strongly correlated compounds (*55*).

In order to illuminate the concept of a spin-Q glass, we qualitatively illustrate the energy landscape in Q-space images for a spin-Q glass in comparison to a ferromagnet (Fig. 2). A long-range ordered state can be related to a global minimum in Q space (*28, 56, 57*), where a single-domain ferromagnetic state is equivalent to a Q = 0 global minimum (Fig. 2A). In contrast, a spin-Q glass is distinguished by the existence of flat valleys defined by a distribution of many local minima, i.e. Q-pockets, at finite Q values (Fig. 2B). Broad Q-pockets led to a lack of a preferentially long-range ordered state. Instead, there were

local regions defined by strong local order derived from a spectral weight of mixed Q vectors within the given pockets, and different regions exhibited random distributions of this spectral weight (see color image in Fig. 2B). The superposition of this spatially varying magnetization led to an overall broadening in the spectral weight. Note that multi-Q states for thin 3*d* transition metal films show well-ordered domains (*58*). Within the concept of self-induced glassiness for spins, such pockets may result from a strong competition of magnetic interactions, leading to highly degenerate states (*15, 16*).

Theoretical analysis of the magnetic landscape of Nd(0001)

In order to analyze the origin of the spin-Q glass state in Nd(0001) and its unexpected magnetic patterns (*59*), we used ab initio calculations to quantify and understand the exchange interactions and the energy landscape in Q space of bulk Nd, which adopts a dhcp structure that is critical for its magnetic exchange interactions (Fig. 3A). The RSPt code was used for this purpose (see methods and supplemental material, section S4) (*50*). The calculated exchange interactions for bulk Nd are shown for both the dhcp and a hypothetical hcp structure and note the minute energy difference between these crystal structures, resulting from the large similarities in atomic arrangement.

The distance dependence of the exchange interactions in the hcp structure illustrates a prototypical behavior with ferromagnetic nearest neighbor interactions and an oscillating Ruderman-Kittel-Kasuya-Yosida (RKKY)–like interaction at large distances, as seen with other lanthanides such as Gd (*60, 61*). In contrast, in the dhcp, at shorter range, the interactions are much weaker and primarily antiferromagnetic, but at larger distances, an RKKY interaction sets in. These strong competing magnetic interactions in Nd create conditions for frustrated magnetism and spin-glass formation, as described in the picture of self-induced glassiness (*15*-*17*). Calculated values of the magnetic moment of the surface atoms, and for deeper layers, were similar to the bulk value. The bulk moment was restored within three layers beneath the surface (supplementary text, section S4) (*50*).

In order to clarify the impact of the calculated exchange interactions in the dhcp structure of Nd, we evaluated the magnetic energy landscape by means of single-Q spin spirals, i.e. magnetic structures that

can be parametrized by a single wave vector. For this purpose, we calculated the energy of helical spin spirals. Using the calculated magnetic exchange interactions from Fig. 3A, we parametrized an effective Heisenberg spin Hamiltonian from which the energy $E(\mathbf{Q})$ of the single-Q spirals was then calculated (Fig. 3B). The energy-landscape exploration was performed by fixing the *z*-component Q_z and then sweeping over Q_x and Q_y in the first Brillouin zone (BZ) (supplementary text, section S6) (*50*).

In Fig. 3B, we present the single-Q energy landscape for all possible Q_x and Q_y combinations in the cell spanned by the dhcp reciprocal lattice vectors for the case when $Q_z = 2\pi/c$, which corresponded to the configuration with the lowest single-Q energy. This choice of Q_z corresponded to a 90° rotation of the moments in adjacent atomic layers within the dhcp unit cell. The Q-dependent energy is color-coded such that red (blue) regions correspond to spin spirals with low (high) energy. This visualization illustrates the complex energy landscape of Nd, as exemplified by the broad and flat dark-red ring-like structure, showing similarity to the deduced landscape in (*26*).

Instead of a distinct, six-fold degenerate set of strong energy minima that could be expected for a spin-spiral magnet on a hexagonal lattice, the red ring structure showed that the energy barriers between global and local minima in this region of the BZ were very small. In addition, high-energy local minima, or pockets (in red) were distributed with hexagonal symmetry, just inside the BZ, and another low-energy valley-like structure formed along the BZ boundary. Magnetic anisotropy was not considered here, and we expect that it would bias the ring structure toward the high-symmetry directions, creating pockets akin to those seen in the experiments. Thus, Fig. 3B illustrates that the energy landscape of Nd has several broad Q-pockets, which supports the formation of the experimentally observed spin-Q glass structure. The presence of strongly competing interactions leading to a glass-like energy landscape in Q space is a key manifestation of the concept of self-induced glassiness (*13-15*).

Spin-Q glass dynamics: autocorrelation and static correlation function

In addition to the single-Q energy-landscape explorations, we also used Monte Carlo and atomistic spin dynamics (ASD) simulations to find the ground-state magnetic structure. Although not all conventional

spin glasses exhibit identical relaxation dynamics, a common trait is aging, i.e. that the relaxation process slows down over time and never settles on a single equilibrium state. Aging is observed in the Edwards-Anderson model (*1*). In order to characterize the aging dynamics of dhcp Nd we studied the two-time autocorrelation function defined as $C(t_w, t)=\langle \boldsymbol{m}_i(t + t_w) \cdot \boldsymbol{m}_i(t_w)\rangle$, where the brackets denote averaging over all sites of the system. If a system relaxes to a fixed ground state following non-glassy dynamics, the autocorrelation function should increase with increasing waiting times t_w. Autocorrelation analysis based on ASD has earlier been used to capture the multiple relaxation scales of a conventional spin glass system (*10*). This approach, which is typically analyzed with a mean-field approach, is well suited to characterize the multiple time scales indicative of aging (supplementary text, section S8) (*50*).

In Fig. 3C, we show the autocorrelation at logarithmically spaced waiting times t_w for dhcp Nd as it relaxed from a fully disordered state at T = 1 K. The autocorrelation function decayed exponentially towards zero, a typical feature of aging and spin-glass behavior (Fig. 3C) (*50*), and was similar to that of a traditional spin-glass system, like Cu-Mn or that of the Edwards-Anderson model (see supplementary text, section S8) (*10, 50*). In other words, the relaxation process of Nd never arrived at a well-defined energy minimum. We note the autocorrelation analysis often assumes a mean-field picture of aging, whereas a more complete picture should include the Q-dependent contributions to the multiple time scales. These simulations were performed for bulk dhcp Nd meaning that the observed spin-Q glassy behavior can only be a result of the exchange interactions of the system since neither surface states nor defects were present in the simulations.

In addition to mapping out the aging dynamics of the spin-Q glass state, from these simulations, we obtained real-space spin structures (supplementary text, section S7) (*50*) as well as the static correlation function $S(\mathbf{Q})$, which can be compared directly with the experimental Q-space images. In Fig. 3D, we present the simulated peaks of $S(\mathbf{Q})$ in comparison with (i) the observed range of spectral weight from the Q-pockets Q_A, Q_B and Q_C in the SP-STM experiments (B_z = 0, for both pristine zero-field cooled samples and samples after being exposed to magnetic field), and (ii) reported neutron diffraction data (*21, 25*). In comparison, the SP-STM and neutron diffraction data agreed well with the simulated results in regions of

lower Q values. This agreement indicated that the surface-derived measurements were significantly coupled to the bulk magnetic properties. The simulations also provide distinct local maxima of the correlation function, which so far have not been detected experimentally.

Impact of impurities on Q-state distribution

Nd films illustrate characteristics of bulk Nd with the dhcp structure, including the expected surface state of Nd(0001) and the expected strain-free lattice constant (*50*). Although we did not observe bulk screw dislocations and large-scale defects for the island samples, the surface did show the presence of surface impurities. The presence of the interface did not seem to influence the imaged magnetism, e.g. underlying substrate step edges (supplementary text, section S2) (*50*). Before discussing the role of the impurities on the magnetic order, we note that the main concentration of impurities in the source material is oxygen and carbon, each about 2000 parts per million by atom (ppma) (Table S1) (*50*). The impurity density of our surfaces is ca. 0.01 ML, which is below the detection limit of any surface-averaging spectroscopic technique and among the best reported for lanthanide metal surfaces (*53, 62*). Of these impurities, we observe the unperturbed intrinsic width of the surface state (*54*). As the quality of the surface state (i.e. its intensity and width) in spatially averaging photoemission has been demonstrated to illustrate the structural order as well as cleanliness of the surface (*31, 32, 52*), we conclude that the surface defects in the present limit have a negligible impact on the overall electronic structure.

In Fig. 4, we show a comparison between two separately prepared samples and their respective FFTs, in which the defect density is sufficiently different. For samples with higher defect densities, imaged at T = 1.3 K, we observed more well-defined Q vectors at higher overall Q values, with a narrower distribution in Q space compared to samples where the defect density is lower (e.g. Fig. 1). Although local order in this case had a well-defined periodicity, the orientation was not perfectly aligned leading to a small angular distribution of Q states. In stark contrast, as the defect density was reduced, the distribution of Q states strongly broadened. In other words, with cleaner samples, there was a stronger manifestation of multiple energy minima as more Q states stabilize. This observation is in contrast to typical magnetically ordered systems, where any collinear or non-collinear configuration narrows its distribution in reciprocal space

with cleaner samples. In this context, extend line defects (screw and edge dislocations) in lanthanide films were shown to pin magnetic order, while local point defects such as atomic adsorbates did not (*51*). As shown in Fig. S1 and S3 (*50*), thick closed Nd films exhibit line defects as well as higher surface defect densities than the typical islands we show in Fig. 1. The unavoidable presence of dislocations and the higher amount of impurities in bulk single crystals may lead to defect-induced pinning of Q-vectors, exemplifying the importance of cleanliness in order to observe the glassy behavior. However, for the ~100 ML thick closed films we observed qualitatively the same magnetic multi-Q structure as the islands (Fig. S3) (*50*). Hence, despite their seemingly small volume the islands reflect the magnetic properties of bulk Nd. We conclude that below a critical defect density, a multi-well landscape emerged as a spin-Q glass phase, as illustrated in the defect-free calculations of both the energy landscape and the autocorrelation function, in Fig. 3C.

Magnetic field evolution of the spin-Q glass state

We experimentally characterized the response of the magnetization to external magnetic fields. Out-of-plane field dependence is illustrated in Fig. 5 for a few chosen fields up to B_z = 7 T at T = 1.3 K (see also supplementary text, section S9) (*50*). The application of magnetic field should favor states with smaller Q, eventually leading to a preferential Q state near the zone center (*56, 63*), when the Zeeman energy exceeds the local exchange energy. However, magnetic imaging of a given area at variable magnetic field revealed a different picture. At increasing magnetic fields, no distinct Q state became favorable, with the spectral weight strongly broadening along high-symmetry directions toward higher Q (Fig. 5G). The magnetic order was sensitive to fields on the order of B_z = 0.5 T, illustrating the degeneracy driven by the Q-pockets, whereas at the highest applied fields (B_z = 7 T) there was no distinct Q state, demonstrating the strong local exchange energy.

Moreover, the application of in-plane fields revealed similar behavior: favorable Q states collapse onto an axis related to the direction of the applied field, but with appreciable smearing of the spectral weight along one particular axis, reminiscent of the so-called archipelago phase seen in neutron scattering (*22-24*) (Fig. S13) (*50*). The field-dependent behavior seen here cannot be attributed to the picture of local

domains, as there were neither clear domain boundaries that could be traced in magnetic field, nor a repeating or favorable Q-structure. We further note that magnetostriction effects should saturate beyond 1 T, at the measured temperature, as previously reported (*23*).

Aging of the spin-Q glass state through magnetic field

Although field-dependent imaging illustrated the degeneracy within the Q-pockets, the distinguishing property of a spin glass is the observation of aging. Aging, in a mean-field picture, can be described by the existence of multiple relaxation time scales, as exemplified in Fig. 3C, leading to a magnetization state that never fully relaxes (*10*). To monitor the evolution of the ground state, we repeatedly ($i = 1, \dots, n$) apply the following procedure, starting from the pristine ($i = 0$) state : (i) sweep up the magnetic field to $B_{z,i}$ (max. 7 T) and stay at that value for a finite time τ_i, (ii) reduce the magnetic field to zero, (iii) image the i^{th} zero-field state of the same area as the pristine state. Comparing several field-dependent cycles, the zero-field magnetic state at T = 1.3 K illustrates sequential redistributions of the spectral weight within and between the various Q-pockets (Fig. 6).

Consistent with aging, the magnetic state did not relax to a given ground state after perturbation, nor was there a clear tendency toward a different but particular ground state distribution. Instead, the overall trend was a redistribution between all pockets toward a broader overall distribution along the high-symmetry directions, with the angular distribution sharpening along the high-symmetry axes. Moreover, the system never reverted to the initial zero-field cooled state. These findings ruled out that field-dependent cycling can be understood as the evolution of a favorable domain. To rule out hysteretic effects, we also performed field sweeps at positive and negative fields, but there was no clear correlation between such subsequent conditions, nor any correlation with local defects on the surface. We note that similar aging effects, and magnetic field evolution, were seen at different temperatures (Fig. S15) (*50*), as we detail below, as well as with the application of an in-plane field and also near island edges (Fig. S14, see also supplementary text, section S10 and S11) (*50*).

Multiple and Q-dependent relaxation times at elevated temperature

The evolution of intermittent spectral weight distributions, which were frozen after subsequent field-sweep cycles, is a signature of slow aging dynamics and a hierarchy of states separated by small energies. In contrast, the application of temperature illustrated the presence of fast dynamics concomitant with slow relaxation dynamics. To illustrate this effect, we show the effect of temperature by imaging the same area at different temperature (Fig. 7). Compared to T = 1.3 K (Fig. 7B), the spectral weight within the Q_A pockets "melted" away at T = 4.2 K (Fig. 7D). Also in the real-space image, the associated long-wavelength pattern was no longer visible (Fig. 7C) when compared to images of the identical area at T = 1.3 K (Fig. 7A). Our time resolution was limited to roughly 1 ms so that we were imaging the time-averaged magnetization. Thus, the loss of spectral intensity in the Q_A pockets was most likely caused by increased fluctuations of the magnetization states within the Q_A pockets, which were thermally activated at this temperature. Thus, the fluctuations of the magnetization lead to an overall reduction of the measured time-averaged spin polarization. Nevertheless, we observed similar slow aging behavior after application of magnetic field at T = 4.2 K for the other Q-pockets as seen at lower temperatures for exactly the same sample area (Fig. 7F). This observation illustrated that there were at least two different and Q-dependent time scales present at sufficient temperature.

Although it is unclear how to describe the Q-dependent dynamical behavior of the magnetization, the presence of multiple relaxation times is a clue of glassy behavior in a mean-field picture, analogous to aging effects seen in metallic alloys (*3, 10*). We note that there may be some ruggedness to the energy barriers, symbolic of a multi-well landscape, resulting from strong local order in this system. In structural glasses, local order leads to a more complex picture of the dynamics, referred to as dynamic heterogeneity (*64*). In this picture, there can be different spatial regions exhibiting dramatically different dynamics related to the local correlations. The existence of different and Q-dependent time scales suggests evidence for dynamic heterogeneity in Nd. Therefore, it is not entirely clear how many various time scales can be seen in this system, and at what temperatures, compared to conventional spin glasses which have more flat energy landscapes. Combined with the observations that lower defect densities led to a smearing of the Q states, these results exemplify the complex aging behavior related to the multi-well energy landscape illustrated by theory in Fig. 3. The application of higher temperature led to more well-

defined Q states (e.g. for T = 7 K); the same area imaged at T = 40 mK, in comparison, shows similar glassy distributions to the previously shown data taken at T = 1.3 K (supplementary text, section S11) (*50*). This result rules out transitions into various long-range ordered multi-Q states (*21, 25*) with decreasing temperature, but rather exemplifies the emergence of a spin-Q glass state.

Discussion

We have demonstrated that the magnetic ground state of elemental Nd is a spin-Q glass. We have observed aging, that is, glassiness, in an elemental magnetic solid with minimal amounts of chemical or extrinsic disorder, which is direct experimental confirmation of self-induced glassiness (*15*). Moreover, the coexistence of short-range order exhibited by multi-Q pockets along with aging behavior in a material without extrinsic disorder cannot be captured by traditional mean-field theoretical descriptions of spin glasses (*1, 2*). The self-induced glassy behavior observed here is a departure from the traditional magnetic alloys where disorder drives the glassy dynamics and likewise many intermediate time scales can be observed. The energy landscape of Nd may possess some amount of ruggedness, resulting from the strong local Q order (*17*), providing a novel material system to study dynamic heterogeneity in a spin glass material (*64*). With the advent of techniques that can probe picosecond dynamics with STM (*65*), it may be interesting to develop magnetic sensitive time-resolved methods that can resolve the picosecond dynamics in neodymium, which can be directly compared to ASD simulations. It remains to be understood what is particularly special about the interplay between crystal structure and electronic properties in Nd that leads to this exotic behavior, necessitating a deeper theoretical understanding of the role of electron correlation effects, the possible influence of the surface, as well as the interplay between spin and orbital degrees of freedom.

The conclusions drawn here resolve the long-standing debate about the magnetic state of Nd, in that the various reported multi-Q transitions as a function of temperature can be understood within a picture of multiple minima in Q space with different depths that can be thermally activated. Likewise, the establishment of spin-Q glass order raises the question of the dynamical behavior of spins through the variation in local relaxation times, and may provide a material platforms to explore exotic topological

quasiparticles similar to fractons (*66-68*). The example here expands our views on magnetic states of matter, necessitating numerous further experimental and theoretical investigation in order to understand the emergence of aging behavior in magnetic systems.

Methods

The experimental studies were performed in two home-built ultrahigh vacuum (UHV) systems that allow for cleaning the W(110) single crystal substrates, molecular beam epitaxy and annealing of Nd (see supplementary text, section S1) as well as transfer into the cryogenic STM, all in situ. The first system operates at a base temperature of 1.2 K with magnetic fields up to 9 T perpendicular to the sample. The second system operates at a base temperature of 30 mK by with magnetic fields up to 9 T perpendicular and 4 T parallel to the sample (*69*). The SP-STM measurements were performed using an antiferromagnetic Cr tip with out-of-plane spin polarization (see supplementary text, section S2). Apart from a global plane subtraction, all STM topography images are unprocessed raw data. Magnetization images were produced by subtraction of the majority and the minority SP-STM images, and corresponding Q-space images were produced by computing the FFT using MATLAB. For the latter, we again did not apply any post-processing (see supplementary text, section S2).

The theoretical studies included ab initio calculations of bulk Nd as well as cubic- and hexagonal-terminated slabs with up to 13 layers. The electronic structure was calculated within density functional theory by means of the RSPt software (*70*). We used the local spin-density approximation for the exchange and correlation functional. In these calculations we made use of the standard model of the lanthanides and treated the 4*f* electrons as localized, unhybridized with a magnetic moment according to Russel-Saunders coupling. The dispersive states were expanded by means of 6*s*, 6*p* and 5*d* orbitals in a multiple basis fashion, as described in (*70*). In addition, we included pseudo-core 5*s* and 5*p* states to hybridize with the rest of the valence states. No approximation was made concerning the shape of the charge density or potential, in a so-called full-potential description. The simulations were done using 32768 and 1152 k-points of the full Brillouin-zone for bulk dhcp and slabs, respectively (see also

supplementary text, section S4). The MC/ASD calculations were performed using the UppASD software (*71, 72*) (see supplementary text, section S5).

References and Notes

1. S. F. Edwards, P. W. Anderson, Theory of spin glasses. *J Phys F: Met Phys* **5**, 965 (1975).
2. S. F. Edwards, P. W. Anderson, Theory of spin glasses. II. *J Phys F: Met Phys* **6**, 1927 (1976).
3. K. Binder, A. P. Young, Spin glasses: Experimental facts, theoretical concepts, and open questions. *Rev Mod Phys* **58**, 801 (1986).
4. M. Mezard, G. Parisi, M. A. Virasoro, *Spin Glass Theory and Beyond*. (World Scientific, Singapore, 1987).
5. K. H. Fischer, J. A. Hertz, *Spin Glasses*. (Cambridge University Press, 1993).
6. N. Katayama *et al.*, Investigation of the Spin-Glass Regime between the Antiferromagnetic and Superconducting Phases in Fe1+ySexTe1-x. *J Phys Soc Jap* **79**, 113702 (2010).
7. J. Hertz, A. Krough, R. G. Palmer, *Introduction to the theory of neural computation*. (CRC Press, 1991).
8. Y. Zhou, K. Kanoda, T.-K. Ng, Quantum spin liquid states. *Rev Mod Phys* **89**, 025003 (2017).
9. S. T. Bramwell, M. J. P. Gingras, Spin Ice State in Frustrated Magnetic Pyrochlore Materials. *Science* **294**, 1495 (2001).
10. B. Skubic *et al.*, Atomistic spin dynamics of the Cu-Mn spin-glass alloy. *Phys Rev B* **79**, 024411 (2009).
11. R. Rammal, G. Toulouse, M. A. Virasoro, Ultrametricity for physicists. *Rev Mod Phys* **58**, 765 (1986).
12. M. Mézard, G. Parisi, A tentative replica study of the glass transition. *J Phys A: Mat Theor* **29**, 6515-6524 (1996).
13. J. Schmalian, P. G. Wolynes, Stripe Glasses: Self-Generated Randomness in a Uniformly Frustrated System. *Phys Rev Lett* **85**, 836 (2000).
14. H. Westfahl, J. Schmalian, P. G. Wolynes, Self-generated randomness, defect wandering, and viscous flow in stripe glasses. *Phys Rev B* **64**, 174203 (2001).
15. A. Principi, M. I. Katsnelson, Self-Induced Glassiness and Pattern Formation in Spin Systems Subject to Long-Range Interactions. *Phys Rev Lett* **117**, 137201 (2016).
16. A. Principi, M. I. Katsnelson, Stripe glasses in ferromagnetic thin films. *Phys Rev B* **93**, 054410 (2016).
17. A. Kolmus, M. I. Katsnelson, A. A. Khajetoorians, H. J. Kappen, Atom-by-atom construction of attractors in a tunable finite size spin array. *New J Phys* **22**, 023038 (2020).
18. R. M. Moon, W. C. Koehler, W. Cable, Magnetic Structure of Neodymium. *J Appl Phys* **35**, 1041 (1964).
19. P. Bak, B. Lebech, Triple-Q-Modulated Magnetic-Structure and Critical Behavior of Neodymium. *Phys Rev Lett* **40**, 800 (1978).
20. B. Lebech, J. Alsnielsen, K. A. Mcewen, X-Ray and Neutron-Scattering Study of the Magnetic-Structure of Neodymium Metal. *Phys Rev Lett* **43**, 65 (1979).
21. E. M. Forgan, E. P. Gibbons, K. A. Mcewen, D. Fort, Observation of a Quadruple-Q Magnetic-Structure in Neodymium. *Phys Rev Lett* **62**, 470 (1989).
22. K. A. McEwen, S. W. Zochowski, Magnetic Phase-Transitions in Neodymium. *J Magn Magn Mater* **90-1**, 94 (1990).
23. S. W. Zochowski, K. A. Mcewen, E. Fawcett, Magnetic Phase-Diagrams of Neodymium. *J Phys. Condens. Matter* **3**, 8079 (1991).
24. E. M. Forgan *et al.*, Field Effects on the Antiferromagnetic Ordering of Neodymium. *J Magn Magn Mater* **104**, 911 (1992).

25. B. Lebech, J. Wolny, R. M. Moon, Magnetic Phase-Transitions in Double Hexagonal Close-Packed Neodymium Metal Commensurate in 2 Dimensions. *J Phys. Condens. Matter* **6**, 5201 (1994).
26. P. A. Lindgard, T. Chatterji, K. Prokes, V. Sikolenko, J. U. Hoffmann, Magnetic diffuse scattering from Nd above T-N and deduced exchange interaction parameters. *J Phys. Condens. Matter* **19**, 286201 (2007).
27. D. Wegner, A. Bauer, A. Rehbein, G. Kaindl, Exchange Splittings of Lanthanide (0001)-Surface States and Their Dependences on Short-Range Magnetic Order. *Jap J Appl Phys* **45**, 1941 (2006).
28. S. Heinze *et al.*, Spontaneous atomic-scale magnetic skyrmion lattice in two dimensions. *Nat Phys* **7**, 713 (2011).
29. D. Weller, S. F. Alvarado, Preparation of Remanently Ferromagnetic Gd(0001). *J Appl Phys* **59**, 2908 (1986).
30. S. C. Wu *et al.*, Surface-State on Clean Tb(0001). *Phys Rev B* **44**, 13720 (1991).
31. G. H. Fecher *et al.*, Dichroism in angular resolved VUV-photoemission from the (0001) surfaces of thin Gd and Nd films epitaxially grown on W(110). *Eur Phys J B* **11**, 161 (1999).
32. E. Weschke, G. Kaindl, 4f- and surface-electronic structure of lanthanide metals. *J Electron Spectros Relat Phenomena* **75**, 233 (1995).
33. K. M. Dobrich *et al.*, Fermi-Surface Topology and Helical Antiferromagnetism in Heavy Lanthanide Metals. *Phys Rev Lett* **104**, 246401 (2010).
34. C. Dufour *et al.*, Epitaxial growth of dhcp samarium: single crystal films and Sm/Nd superlattices. *J Cryst Growth* **234**, 447 (2002).
35. O. Waller *et al.*, Lattice dynamics of neodymium: Influence of 4 f electron correlations. *Phys Rev B* **94**, 014303 (2016).
36. E. Weschke, G. Kaindl, Magnetic exchange splitting in lanthanide metals. *J Phys. Condens. Matter* **13**, 11133 (2001).
37. J. Quinn, Y. S. Li, F. Jona, D. Fort, Atomic-Structure of a Gd(0001) Surface. *Phys Rev B* **46**, 9694 (1992).
38. J. P. Goff, C. Bryn-Jacobsen, D. F. McMorrow, R. C. C. Ward, M. R. Wells, Formation of uniform magnetic structures and epitaxial hydride phases in Nd/Pr superlattices. *Phys Rev B* **55**, 12537 (1997).
39. E. D. Tober, R. X. Ynzunza, C. Westphal, C. S. Fadley, Relationship between morphology and magnetic behavior for Gd thin films on W(110). *Phys Rev B* **53**, 5444 (1996).
40. S. Jo, Y. Gotoh, Growth modes of Nd deposited on Mo(110) surface. *Jap J Appl Phys* **38**, 6878 (1999).
41. H. Li *et al.*, Structural and Electronic-Properties of Ultrathin Films of Gd, Tb, Dy, Ho, and Er. *Phys Rev B* **45**, 3853 (1992).
42. M. Farle, K. Baberschke, U. Stetter, A. Aspelmeier, F. Gerhardter, Thickness-Dependent Curie-Temperature of Gd(0001)/W(110) and Its Dependence on the Growth-Conditions. *Phys Rev B* **47**, 11571 (1993).
43. C. S. Arnold, D. P. Pappas, Gd(0001): A semi-infinite three-dimensional heisenberg ferromagnet with ordinary surface transition. *Phys Rev Lett* **85**, 5202 (2000).
44. C. Schussler-Langeheine *et al.*, Magnetic splitting of valence states in ferromagnetic and antiferromagnetic lanthanide metals. *Phys Rev Lett* **84**, 5624 (2000).
45. E. Weschke *et al.*, Finite-size effect on magnetic ordering temperatures in long-period antiferromagnets: Holmium thin films. *Phys Rev Lett* **93**, 157204 (2004).
46. B. A. Everitt *et al.*, Helimagnetic structures in epitaxial Nd/Y superlattices and alloys. *Phys Rev B* **56**, 5452 (1997).
47. J. P. Goff *et al.*, Magnetism and superconductivity in neodymium/lanthanum superlattices. *Physica B* **241**, 714 (1998).
48. A. Aspelmeier, F. Gerhardter, K. Baberschke, Magnetism and Structure of Ultrathin Gd Films. *J Magn Magn Mater* **132**, 22 (1994).
49. M. Bode, M. Getzlaff, S. Heinze, R. Pascal, R. Wiesendanger, Magnetic exchange splitting of the Gd(0001) surface state studied by variable-temperature scanning tunneling spectroscopy. *Appl Phys A* **66**, S121 (1998).
50. S. s. materials, See supplementary materials. *See supplementary materials*.

51. S. Krause *et al.*, Consequences of line defects on the magnetic structure of high anisotropy films: Pinning centers on Dy/W(110). *Europhys Lett* **76**, 637 (2006).
52. M. Getzlaff, M. Bode, R. Pascal, R. Wiesendanger, Adsorbates on Gd(0001): A combined scanning tunneling microscopy and photoemission study. *Phys Rev B* **59**, 8195 (1999).
53. D. Wegner, A. Bauer, G. Kaindl, Effect of impurities on Tamm-like lanthanide-metal surface states. *Phys Rev B* **76**, 113410 (2007).
54. D. Wegner, A. Bauer, G. Kaindl, Magnon-broadening of exchange-split surface states on lanthanide metals. *Phys Rev B* **73**, 165415 (2006).
55. T. Hanaguri *et al.*, A 'checkerboard' electronic crystal state in lightly hole-doped Ca2-xNaxCuO2Cl2. *Nature* **430**, 1001 (2004).
56. S. Muhlbauer *et al.*, Skyrmion Lattice in a Chiral Magnet. *Science* **323**, 915 (2009).
57. X. Z. Yu *et al.*, Real-space observation of a two-dimensional skyrmion crystal. *Nature* **465**, 901 (2010).
58. K. von Bergmann, M. Menzel, A. Kubetzka, R. Wiesendanger, Influence of the Local Atom Configuration on a Hexagonal Skyrmion Lattice. *Nano Lett* **15**, 3280 (2015).
59. W. C. Koehler, Magnetic Properties of Rare-Earth Metals and Alloys. *J Appl Phys* **36**, 1078 (1965).
60. I. L. M. Locht *et al.*, Standard model of the rare earths analyzed from the Hubbard I approximation. *Phys Rev B* **94**, 085137 (2016).
61. Y. O. Kvashnin *et al.*, Exchange parameters of strongly correlated materials: Extraction from spin-polarized density functional theory plus dynamical mean-field theory. *Phys Rev B* **91**, 125133 (2015).
62. P. Loptien, L. Zhou, A. A. Khajetoorians, J. Wiebe, R. Wiesendanger, Superconductivity of lanthanum revisited: enhanced critical temperature in the clean limit. *J Phys. Condens. Matter* **26**, 425703 (2014).
63. N. Romming *et al.*, Writing and Deleting Single Magnetic Skyrmions. *Science* **341**, 636 (2013).
64. M. D. Ediger, Spatially Heterogeneous Dynamics in Supercooled Liquids. *Annu Rev Phys Chem* **51**, 99-128 (2000).
65. T. L. Cocker, D. Peller, P. Yu, J. Repp, R. Huber, Tracking the ultrafast motion of a single molecule by femtosecond orbital imaging. *Nature* **539**, 263 (2016).
66. H. Rieger, A. P. Young, in *Complex Behaviour of Glassy Systems,* M. Rubí, C. Pérez-Vicente, Eds. (Springer, Heidelberg, 1997).
67. C. Chamon, Quantum glassiness in strongly correlated clean systems: An example of topological overprotection. *Phys Rev Lett* **94**, 040402 (2005).
68. R. M. Nandkishore, M. Hermele, Fractons. *Annu Rev Condens Matter Phys* **10**, 295 (2019).
69. H. von Allwörden *et al.*, Design and performance of an ultra-high vacuum scanning tunneling microscope operating at 30 mK and in a vector magnetic field. *Rev Sci Instr* **89**, 033902 (2018).
70. J. M. Wills, Alouani, M., Andersson, P., Delin, A., Eriksson, O., Grechnyev, O. , *Full-Potential Electronic Structure Method*. (Springer, Heidelberg, 2010).
71. https://github.com/UppASD/UppASD.
72. O. Eriksson, A. Bergman, L. Bergqvist, J. Hellsvik, *Atomistic Spin Dynamics: Foundations and Applications*. (Oxford University Press, 2017).
73. B. Kim, A. B. Andrews, J. L. Erskine, K. J. Kim, B. N. Harmon, Temperature-Dependent Conduction-Band Exchange Splitting in Ferromagnetic Hcp Gadolinium - Theoretical Predictions and Photoemission Experiments. *Phys Rev Lett* **68**, 1931 (1992).
74. M. E. Davila, S. L. Molodtsov, C. Laubschat, M. C. Asensio, Structural determination of Yb single-crystal films grown on W(110) using photoelectron diffraction. *Phys Rev B* **66**, 035411 (2002).
75. S. A. Nepijko *et al.*, Lattice relaxation of Gd on W(110). *Surf Sci* **466**, 89 (2000).
76. H. Ott, C. Schussler-Langeheine, E. Schierle, E. Weschke, G. Kaindl, Depth-resolved magnetic structure across the ferromagnetic to helical-antiferromagnetic phase transition in Dy/W(110). *Phys Rev B* **82**, 214408 (2010).
77. S. Soriano, C. Dufour, K. Dumesnil, A. Stunault, New magnetic phase and magnetic coherence in Nd/Sm(001) superlattices. *J Phys. Condens. Matter* **18**, 4995 (2006).
78. E. D. Tober *et al.*, Observation of a ferromagnetic-to-paramagnetic phase transition on a ferromagnetic surface using spin-polarized photoelectron diffraction: Gd(0001). *Phys Rev Lett* **81**, 2360 (1998).

79. A. Bauer, A. Muhlig, D. Wegner, G. Kaindl, Lifetime of surface states on (0001) surfaces of lanthanide metals. *Phys Rev B* **65**, 075421 (2002).
80. L. Berbil-Bautista, S. Krause, M. Bode, R. Wiesendanger, Spin-polarized scanning tunneling microscopy and spectroscopy of ferromagnetic Dy(0001)/W(110) films. *Phys Rev B* **76**, 064411 (2007).
81. M. Bode *et al.*, Temperature-dependent exchange splitting of a surface state on a local-moment magnet: Tb(0001). *Phys Rev Lett* **83**, 3017 (1999).
82. M. Bode, M. Getzlaff, R. Wiesendanger, Spin-polarized vacuum tunneling into the exchange-split surface state of Gd(0001). *Phys Rev Lett* **81**, 4256 (1998).
83. M. Bodenbach, A. Hohr, C. Laubschat, G. Kaindl, M. Methfessel, Surface Electronic-Structure of Tm(0001) and Yb(111). *Phys Rev B* **50**, 14446 (1994).
84. M. Donath, B. Gubanka, F. Passek, Temperature-dependent spin polarization of magnetic surface state at Gd(0001). *Phys Rev Lett* **77**, 5138 (1996).
85. B. A. Everitt *et al.*, Magnetic-Ordering of Nd in Nd/Y Superlattices. *J Magn Magn Mater* **140**, 769 (1995).
86. F. Heigl *et al.*, Annealing-induced extension of the antiferromagnetic phase in epitaxial terbium metal films. *Phys Rev B* **72**, 035417 (2005).
87. E. Lundgren *et al.*, Geometry of the valence transition induced surface reconstruction of Sm(0001). *Phys Rev Lett* **88**, 136102 (2002).
88. K. Maiti, M. C. Malagoli, A. Dallmeyer, C. Carbone, Finite temperature magnetism in Gd: Evidence against a Stoner behavior. *Phys Rev Lett* **88**, 167205 (2002).
89. A. Melnikov *et al.*, Picosecond magnetization dynamics of the Gd(0001) surface. *J Magn Magn Mater* **272**, 1001 (2004).
90. C. Ostertag *et al.*, Dichroism in VUV photoemission from the (0001)-surfaces of ultrathin Gd and Nd films on W(110). *Surf Sci* **377**, 427 (1997).
91. H. Realpe *et al.*, Islands as Nanometric Probes of Strain Distribution in Heterogeneous Surfaces. *Phys Rev Lett* **104**, 056102 (2010).
92. A. Rehbein, D. Wegner, G. Kaindl, A. Bauer, Temperature dependence of lifetimes of Gd(0001) surface states. *Phys Rev B* **67**, 033403 (2003).
93. E. Weschke *et al.*, Temperature dependence of the exchange splitting of the surface state on Gd(0001): Evidence against spin-mixing behavior. *Phys Rev Lett* **77**, 3415 (1996).
94. M. Bode, S. Krause, L. Berbil-Bautista, S. Heinze, R. Wiesendanger, On the preparation and electronic properties of clean W(110) surfaces. *Surf Sci* **601**, 3308 (2007).
95. P. Kurz, G. Bihlmayer, S. Blugel, Magnetism and electronic structure of hcp Gd and the Gd(0001) surface. *J Phys. Condens. Matter* **14**, 6353 (2002).
96. A. C. Jenkins *et al.*, The relationship between interlayer spacing and magnetic ordering in gadolinium. *J Phys. Condens. Matter* **12**, 10441 (2000).
97. W. Kohn, Density functional and density matrix method scaling linearly with the number of atoms. *Phys Rev Lett* **76**, 3168 (1996).
98. N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, E. Teller, Equation of State Calculations by Fast Computing Machines. *J Chem Phys* **21**, 1087 (1953).
99. B. Skubic, J. Hellsvik, L. Nordström, O. Eriksson, A method for atomistic spin dynamics simulations: implementation and examples. *J Phys. Condens. Matter* **20**, 315203 (2008).
100. L. Lundgren, P. Svedlindh, P. Nordblad, O. Beckman, Dynamics of the Relaxation-Time Spectrum in a CuMn Spin-Glass. *Phys Rev Lett* **51**, 911 (1983).

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