Thermally induced magnetic order from glassiness in elemental neodymium

Benjamin Verlhac1, Lorena Niggli1, Anders Bergman2, Umut Kamber1, Andrey Bagrov1,2, Diana Iuşan2, Lars Nordström2, Mikhail I. Katsnelson1, Daniel Wegner1, Olle Eriksson2,3 and Alexander A. Khajetoorians1✉

At finite temperatures, fluctuations invariably introduce disorder and are responsible for ultimately destroying ordered phases. Here we present an unusual magnetic transition in elemental neodymium where, with increasing temperature, long-range multiply periodic ‘multi-Q’ magnetic order emerges from a self-induced spin glass. Using temperature-dependent spin-polarized scanning tunnelling microscopy, we characterize the local order of a previously reported spin glass phase, and quantify the emergence of long-range multi-Q order with increasing temperature. We develop two analysis tools that allow us to determine the glass transition temperature from measurements of the spatially dependent magnetization. We compare these observations with atomistic spin dynamics simulations, which reproduce the qualitative observation of a phase transition from a low-temperature spin glass phase to an intermediate ordered multi-Q phase. These simulations trace the origin of the unexpected high-temperature order in weakened frustration driven by temperature-dependent sublattice correlations. These findings constitute an example of order from disorder, and provide a platform to study the rich magnetization dynamics of a self-induced spin glass.

O}

1Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands. 2Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden. 3School of Science and Technology, Örebro University, Örebro, Sweden. ✉e-mail: a.khajetoorians@science.ru.nl

NATURE PHYSICS | VOL 18 | AUGUST 2022 | 905–911 | www.nature.com/naturephysics

At finite temperatures, fluctuations invariably introduce disorder and are responsible for ultimately destroying ordered phases. Here we present an unusual magnetic transition in elemental neodymium where, with increasing temperature, long-range multiply periodic ‘multi-Q’ magnetic order emerges from a self-induced spin glass. Using temperature-dependent spin-polarized scanning tunnelling microscopy, we characterize the local order of a previously reported spin glass phase, and quantify the emergence of long-range multi-Q order with increasing temperature. We develop two analysis tools that allow us to determine the glass transition temperature from measurements of the spatially dependent magnetization. We compare these observations with atomistic spin dynamics simulations, which reproduce the qualitative observation of a phase transition from a low-temperature spin glass phase to an intermediate ordered multi-Q phase. These simulations trace the origin of the unexpected high-temperature order in weakened frustration driven by temperature-dependent sublattice correlations. These findings constitute an example of order from disorder, and provide a platform to study the rich magnetization dynamics of a self-induced spin glass.

O}

1Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands. 2Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden. 3School of Science and Technology, Örebro University, Örebro, Sweden. ✉e-mail: a.khajetoorians@science.ru.nl

NATURE PHYSICS | VOL 18 | AUGUST 2022 | 905–911 | www.nature.com/naturephysics

At finite temperatures, fluctuations invariably introduce disorder and are responsible for ultimately destroying ordered phases. Here we present an unusual magnetic transition in elemental neodymium where, with increasing temperature, long-range multiply periodic ‘multi-Q’ magnetic order emerges from a self-induced spin glass. Using temperature-dependent spin-polarized scanning tunnelling microscopy, we characterize the local order of a previously reported spin glass phase, and quantify the emergence of long-range multi-Q order with increasing temperature. We develop two analysis tools that allow us to determine the glass transition temperature from measurements of the spatially dependent magnetization. We compare these observations with atomistic spin dynamics simulations, which reproduce the qualitative observation of a phase transition from a low-temperature spin glass phase to an intermediate ordered multi-Q phase. These simulations trace the origin of the unexpected high-temperature order in weakened frustration driven by temperature-dependent sublattice correlations. These findings constitute an example of order from disorder, and provide a platform to study the rich magnetization dynamics of a self-induced spin glass.

O
we qualitatively reproduced the two observed magnetic phases below $T_{\text{N}}$ and related this complex behaviour to the competing interactions driven by the two sublattices of the double hexagonal close-packed (dhcp) crystal structure.

The spin–Q glass state of Nd is characterized by the presence of so-called Q pockets$^{19,20}$, which are each defined by a distribution of favourable Q states. The favourability of multiple Q states is driven by the competing exchange interactions, linked to the dhcp structure of Nd (Fig. 1a). The self-induced spin glass behaviour can be measured by magnetization images taken on the Nd(0001) surface using SP-STM (Fig. 1b) (Methods). The spin-glass state can be measured by magnetization images taken on the Nd(0001) surface below $T_{\text{N}}$, including the smallest Q vectors: Scale bars, 10 nm ($g$–$h$). The coloured dots correspond to the marked regions in $e$–$j$. The local periodicity can be extracted using fast Fourier transforms (FFT) of the magnetization images, or so-called Q-space maps. Using the Q-space image (Fig. 2d), we quantified the values of the multi-Q states, which we describe as diamond-like and stripe-like patterns in the magnetization image, respectively. Close-up views of these patterns and their respective multi-Q states are shown in Fig. 2e–f. We emphasize that from the resolution of the FFT (Fig. 2d), the accuracy of the $q$ value is about $\pm 0.07$ nm$^{-1}$, whereas the angular precision is about $\pm 1.4^\circ$. Within this accuracy, we found that the multi-Q state of the diamond-like patterns (Fig. 2g) can be described as a combination of the Q vectors $q_{10}^{\parallel}$ and $q_{0}^{\parallel}$, both with magnitudes of $2.64$ nm$^{-1}$ and oriented along the [1210] and [1210] directions, respectively. The multi-Q state of the stripe-like patterns (Fig. 2h) can be expressed as a combination of three Q vectors: $q_{1}^{\parallel}$, $q_{2}^{\parallel}$, and $q_{3}^{\parallel}$ with magnitudes of $2.41$ nm$^{-1}$ and $2.69$ nm$^{-1}$ and oriented near the [2110] and [1120] directions, respectively, as well as $q_{3}^{\parallel}$ with a magnitude of $5.12$ nm$^{-1}$ and oriented close to the [1210] direction.
Different from the \( Q \) vectors that describe the diamond order, these \( Q \) vectors were not exactly aligned with the crystallographic directions. The angles of the \( Q \) vectors corresponding to the stripe order varied between 115° and 120°. We note that the \( Q \) vectors of the two observed domains presented similar vectors, which led to an apparent blurring/splitting of certain spots when averaging over all domains (Supplementary Fig. 4). We also note that the observed \( Q \) vectors in the multi-\( Q \) phase fell within the \( Q_6 \) and \( Q_8 \) pockets observed for the spin-\( Q \) glass phase described above. After cooling back to \( T = 5.1 \text{ K} \) (Fig. 2a), the long-range ordered multi-\( Q \) phase disappeared and the spin-\( Q \) glass state reappeared with an aged \( Q \) state distribution that, when compared with the pre-annealed sample, exhibited aging. With further temperature cycling (Supplementary Section 2), we observed the same ordered multi-\( Q \) state at higher temperature, without any indications of aging, whereas we always observed aging of the spin-\( Q \) glass states at the lowest temperatures. This observation clearly indicates that there is a glass transition temperature \( (T_c) \) at a temperature below \( T_N \) that separates the glassy state from a well-defined ordered state.

To quantify \( T_c \) and the multi-\( Q \) phase, we measured magnetization images of the same spatial area at several temperatures. The relevant \( Q \)-space images for \( T = 5.1 \text{ K}, 6.6 \text{ K}, 7.5 \text{ K}, 8.9 \text{ K} \) and \( 11 \text{ K} \) are presented in Fig. 3a–c. The corresponding magnetization images can be found in Supplementary Fig. 5. For \( T < 8 \text{ K} \), the observed \( Q \) pockets were extremely sensitive to the given sample temperature. There was a smooth trend with increasing temperature towards fewer and sharper \( Q \) pockets. The lowest- and highest-magnitude \( Q \) spots faded with increasing temperature, eventually leading to the well-defined multi-\( Q \) state. This behaviour can be seen in the temperature evolution of line cuts of the FFT as shown in Fig. 3f. We observed that the \( Q_6 \) pocket intensity vanished while the variance in the \( Q_6 \) and \( Q_8 \) pockets reduced, leading to sharp peaks for \( T > 8 \text{ K} \). The observation of a multi-\( Q \) state with well-defined domains can be seen from \( T = 8.3 \text{ K} \) and remains robust up to the highest measured temperatures (\( T = 15.6 \text{ K} \)). This is consistent with the ‘double-\( q \)’ phase observed in neutron diffraction\(^{26,28,29} \). Furthermore, the \( Q \)-vector values we measured on the surface were in good agreement with the values in the range of 2.54–2.78 nm\(^{-1} \) measured by neutron diffraction for bulk Nd. We note that above 15.6 K we observed an increase in surface contamination along with the diffusion of defects, limiting the temperature range (\( T < 15.6 \text{ K} \)) over which we could image and properly compare.

To quantify the transition from the glass phase to the multi-\( Q \) phase, we developed an analytical sampling method. This is based on the observation that in the spin-\( Q \) glass phase, there is strong local \( Q \) order, but no long-range order. In contrast, the multi-\( Q \) phase exhibits clear long-range order defined by fewer select \( Q \) vectors than the glass phase. To this end, one can analyse sections of
the Q-space maps as a function of temperature, as done in Fig. 3f. However, it is not sufficient to analyse the Q-space maps as a function of temperature, as the development of long-range order is only indirectly related to the changes in the intensity and variance. On the basis of this observation, we developed a method that uses both the real-space and reciprocal-space magnetization, labelled \( M(r) \) and \( M(Q) \), respectively. First, we randomly sampled a given area in \( M(r) \), and extracted the relevant local Q vectors from the corresponding \( M(Q) \) (Supplementary Information). The corresponding \( M(Q) \) of this randomly sampled area was then compared to subsequent randomly sampled areas. On the basis of the statistical sampling, we used the computed Jensen–Shannon divergence of the various distributions produced from the sampled areas and calculated the total Q-state divergence \( \mathcal{D}_Q(T) \) for all the randomly sampled areas at a given temperature. Figure 3g illustrates \( \mathcal{D}_Q(T) \) for all measured temperatures. The value of \( \mathcal{D}_Q(T) \) was maximally one when the sampled distributions of Q vectors were maximally dissimilar, or minimally zero when these distributions were identical. From Fig. 3g, it can be seen that there was a reduction of the value of \( \mathcal{D}_Q(T) \) with increasing temperature and an eventual plateau, consistent with the development of long-range order as a function of temperature that persists above a given temperature \(( T > 8 \text{ K})\). The reduction in \( \mathcal{D}_Q(T) \) can be understood resulting from the fact that a randomly sampled area at higher temperature better represents the total image as long-range order developed. We note that \( \mathcal{D}_Q(T) \) approached a finite non-zero value due the presence of multiple domains and defects in the analysed images. From this analysis, we could extract \( T_c = 8.1 \pm 0.3 \text{ K} \) using linear fitting and analysing where the slope of \( \mathcal{D}_Q(T) \) approached zero. We also utilized a second complementary method, based on the recent development of complexity\(^3\) that is detailed in the Supplementary Information, which yields a similar value of \( T_c (7.9 \pm 0.2 \text{ K}) \).

To further understand the physical origin of the spin-Q glass to multi-Q transition, we performed atomistic spin dynamics and Monte Carlo simulations based on exchange parameters obtained from ab initio electronic structure calculations (for a review, see ref.\(^{11}\)). We simulated the magnetic ground states and thermodynamic properties for a range of temperatures between \( 1 \text{ K} \) and \( 15 \text{ K} \). The specific heat, which was obtained from the derivative of the total energy with respect to temperature\(^1\), is shown in Fig. 4a and exhibits two distinct peaks. As a peak in the specific heat indicates a magnetic phase transition, our spin simulations could thus identify two different phase transitions with increasing temperature. The low-temperature state was identified as the spin-Q glass state in ref.\(^{20}\). An analysis of the two-time autocorrelation function \((\text{Supplementary Section 6})\), which can be extracted from the simulations, confirmed that Nd exhibits the spin-Q glass phase in the whole range of \( T = 0 - 4 \text{ K}\). In contrast, for the intermediate temperature range \( 4 \text{ K} < T < 11 \text{ K} \), the simulations did not indicate any signs of glassy dynamics. The magnetic order in this range was determined by the static correlation function \( S(Q) \) (see Methods for computational details) and could be characterized as a traditional multi-Q phase, as we discuss further below. Above the second predicted phase transition temperature at \( T = 11.5 \text{ K} \), the system becomes paramagnetic. Although there were discrepancies between these precise temperatures and the experimental data, the phase diagram shows strong qualitative agreement with the experimental findings. Namely, below the computed \( T_c (11.5 \text{ K}) \), there is first an ordered phase driven by broken frustration and ultimately a self-induced spin glass phase.

To explain what drives the spin-Q glass behaviour and causes the transition to the multi-Q state, we investigated the role of the two different sublattices present in the dhcp structure. In ref.\(^{20}\), a notable difference between the magnetic interactions for the different sublattice sites was identified. For reference, these interactions have been plotted in Supplementary Fig. 12 and decomposed into the various sublattice contributions. The difference in stacking provided a sufficiently important difference in electronic structure
to produce drastically different exchange values. The interaction between Nd atoms at cubic positions and atoms at other cubic positions (cub–cub in Supplementary Fig. 12) was different from that of Nd atoms at hexagonal positions that interact with other Nd atoms at hexagonal positions (hex–hex in Supplementary Fig. 12). Cross interactions between cubic and hexagonal positions (cub–hex) were also unique, which simply implies that the local symmetry gives unique short- and long-range interactions.

We investigated the sublattice effect further by studying the temporal and spatial correlations between Nd atoms at different sublattice sites. In Fig. 4b–d we show static correlation functions obtained by considering only the correlations between atoms on the same sublattice; that is, atoms at cubic and hexagonal sites in the system. For comparison, we also show the total \( \langle S(Q) \rangle_{\text{total}} \). We observed notable differences between the sublattice correlations: the cubic–cubic sublattice correlations \( \langle S(Q)_{\text{cub-cub}} \rangle \) exhibited not only different maxima but also different temperature behaviour. From our simulations we observed that the cubic–cubic correlations vanished at the first phase transition temperature, whereas the hexagonal–hexagonal correlations prevailed up to the paramagnetic transition. These results can be interpreted as follows: at low temperatures, both sublattice magnetizations have different tendencies towards magnetic order; that is, they would prefer two different magnetic orderings. It is the competition between these ordering tendencies that effectively results in the spin-\( Q \) glass behaviour. The effective strength of the exchange interactions on the cubic sublattice is weaker than the interactions on the hexagonal sublattice, which results in the cubic correlations vanishing at a lower temperature (that is, the first phase transition at \( T=4 \) K). Without the competition from the magnetic atoms on the cubic sublattice, the magnetic order driven by a transition to a paramagnetic phase. When comparing the sublattice decomposition for \( S(Q) \), we observed notable differences between the sublattice correlations: the cubic–cubic sublattice correlations \( \langle S(Q)_{\text{cub-cub}} \rangle \) exhibited not only different maxima than the hexagonal–hexagonal sublattice correlations \( \langle S(Q)_{\text{hex-hex}} \rangle \) but also different temperature behaviour. From our simulations we observed that the cubic–cubic correlations vanished at the first phase transition temperature, whereas the hexagonal–hexagonal correlations prevailed up to the paramagnetic transition. These results can be interpreted as follows: at low temperatures, both sublattice magnetizations have different tendencies towards magnetic order; that is, they would prefer two different magnetic orderings. It is the competition between these ordering tendencies that effectively results in the spin-\( Q \) glass behaviour. The effective strength of the exchange interactions on the cubic sublattice is weaker than the interactions on the hexagonal sublattice, which results in the cubic correlations vanishing at a lower temperature (that is, the first phase transition at \( T=4 \) K). Without the competition from the magnetic atoms on the cubic sublattice, the magnetic order driven by...
the atoms on the hexagonal sites dominates and the result is then a non-glassy multi-Q state that persists up to the paramagnetic transition. The transition from the spin-Q glass phase to the ordered phase at intermediate temperature can also be seen by taking a line cut of $S(Q)$ along $(\Gamma-M)$ (Fig. 4e). An overall broadened intensity can be seen near the experimentally determined $Q_g$ pocket, for $T=1 \text{K}$, which substantially sharpens at $T=6 \text{K}$. We note that in the calculations, only one $Q$ state was observed in the intermediate phase, whereas a multi-Q phase was seen with both neutron diffraction ($T<19.1 \text{K}$)26,27 and our STM-based studies. Although there are deviations in both the precise temperature and the $Q$ values seen between experiments and calculations (Supplementary Section 6), the overall trends of both experimentally observed phases are reproduced in the simulations.

To identify the presence (or lack) of glassy dynamics in the magnetic phases of Nd, we studied the two-time autocorrelation function $C(t, t') = \langle m_i(t) m_i(t') \rangle$, where $m_i$ is the magnetic moment of site $i$, which depends on the simulated time $t$ and the waiting time $t'$. In practice, the simulations start from a random distribution of atomic spins that are allowed to relax according to the atomistic Landau-Lifshitz–Gilbert equation for a time $t_c$. After that the two-time autocorrelation was calculated for a time period $t$. The results of the autocorrelation simulations are shown in Fig. 4f–h, where the results for temperatures $T=1 \text{K}, 6 \text{K}$ and $14 \text{K}$ are presented. At $T=1 \text{K}$ the autocorrelation functions show multiple relaxation times depending on $t_c$, which is a typical signal of spin glass behaviour25. In contrast, at $T=6 \text{K}$, the relaxation behaviour was similar regardless of $t_c$ as with a long-range ordered state. The relaxation behaviour at $T=6 \text{K}$ exhibited different saturation values, which can be explained by the fact that the finite-size simulations performed have no anisotropy or other considerations that keep the system from performing global rotations due to the thermal fluctuations. At $T=14 \text{K}$, the system was paramagnetic and the rapid relaxation rate came from the thermally induced disorder in the simulated system.

Our experimental finding and theoretical analysis partly support, but also expand, the interpretations from neutron diffraction, susceptibility, heat capacity, thermal expansion and other measurements19–21. First, we confirm that there is an ordered multi-Q phase above $T_g \approx 8.1 \text{K}$ (which has been referred to as $T_z$ in the literature), driven by coupling of the hexagonal site spins. Although the neutron diffraction data was interpreted as showing that the multi-Q phase was a relatively simple double-$q$ structure21, our data clearly show that various domains with a total of five different $q$ vectors exist. The fact that these (and their higher harmonic) vectors are very close to each other can easily obscure their presence in reciprocal space when averaging over many domains (as exemplified in Supplementary Fig. 4e). This may explain the interpretation of the neutron diffraction data, and underscores the need for a spatially resolved technique to unravel the complex magnetic structure of Nd. Second, we confirm that the phase transition is driven by the onset of coupling of the cubic site spins. This also illustrates that the surface magnetization pattern, as probed by SP-STM, is representative of the bulk. However, the phase below $8 \text{K}$ was interpreted as a ‘double-$q$’ magnetic structure19,20, we identified it as the glass transition point, below which no long-range order can be found. We note that we did not observe notable changes in the magnetic structure around $6 \text{K}$ in comparison to higher temperatures, whereas previous studies identified another phase transition near $6 \text{K}$ that was interpreted as a ‘quadrupole-$q$’ structure22–24. We merely observed gradual changes in the distribution of, and within, the various $Q$ pockets. The most important effect is the gradual appearance with decreasing temperature of $Q_g$ pockets, as well as pockets at angles far from the crystallographic axes. Although the former was not observed in neutron diffraction studies, the latter could be easily misinterpreted as stemming from a fourth vector. We would like to emphasize that the spin-$Q$ glass phase is very sensitive to even small defect concentrations. In a previous study with a typical surface defect concentration of $\approx 0.010 \text{ML}$ (ref. 26), we found that the $Q_g$ pockets had already ‘melted’ away at $4.2 \text{K}$. In the present data, with four times lower defect concentration, the $Q_g$ pockets were visible up to $T_g$. The defect-induced pinning of $Q$ vectors, leading to ‘less glassiness’, may also account for the deviations found in neutron diffraction experiments.

In conclusion, we have demonstrated the emergence of an ordered phase when the spin-$Q$ glass observed for elemental Nd is heated above $T_g$. This phenomenon is distinct from the debated re-entrant spin glasses1, with the important distinction that the phase observed here is not driven by disorder, a clear coexistence phase or linked to cluster or impurity densities that may influence the magnetism. The origin of this unexpected multi-Q ordering can be traced to quenching of one set of sublattice spin correlations, which is the essential ingredient for driving the spin-$Q$ glass phase. In magnetic systems, it is known that frustration induced by competing interactions can lead to both complicated (for example, noncollinear) magnetic ordering and to spin glassiness. We demonstrate an example of a phase transition between these states and show that the combination of frustration and temperature can lead to very counter-intuitive behaviour, with the emergence of regular and ordered magnetic patterns at relatively higher temperatures. The example of Nd provides a unique platform to investigate complex magnetization dynamics in a system with strong short-range spin correlations that can be tuned by temperature. This could be extended to various other magnetic materials that exhibit temperature-dependent modulations, such as antiferromagnetic-to-ferromagnetic transitions in FeRh (ref. 19), as well as metallic materials that are candidates for fluctuation induced order2, for example PrPtAl (ref. 19). Nd may also provide a platform to study the dynamical behaviour present in glass systems, such as dynamic heterogeneity. This gives a solid experimental and material-specific theoretical background for the concept of order from disorder, which is claimed to be of importance far beyond physics25,26.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01633-9.

Received: 10 September 2021; Accepted: 9 May 2022; Published online: 4 July 2022

References
1. Valasek, J. Piezo-electric activity of Rochelle salt under various conditions. Phys. Rev. 19, 478–491 (1922).
2. Jonasson, K., Mattsson, J. & Nordblad, P. Chaos in the ferromagnetic phase of a reentrant ferromagnet. Phys. Rev. Lett. 77, 2562–2565 (1996).
3. Shimizu, M., Inoue, J. & Nagasawa, S. Electronic structure and magnetic properties of Y-Ni intermetallic compounds. J. Phys. F 14, 2673–2687 (1984).
4. Ballou, R., Gorges, B., Melho, P. & Rouault, P. “Thermal spontaneous magnetization” in Y$_2$Ni$_7$: a misinterpretation. J. Magn. Magn. Mater. 84, L1–L4 (1990).
5. Vonskovsky, S. et al. Temperature induced ferromagnetism-pyromagnetism. J. Phys. Colloq. 49, 253–254 (1988).
6. Henley, C. L. Ordering due to disorder in a frustrated vector antiferromagnet. Phys. Rev. Lett. 62, 2056–2059 (1989).
7. Villain, J., Bidaux, R., Carton, J.-P. & Conte, R. Order as an effect of disorder. J. Phys. F 14, 253–254 (1988).
8. Abdal-Jabbar, G. et al. Modulated magnetism in PrPtAl. Nat. Phys. 11, 321–327 (2015).
9. Mezard, M., Parisi, G. & Virasoro, M. A. Spin Glass Theory and Beyond (World Scientific, 1987).
10. Fischer, K. H. & Hertz, J. A. Spin Glasses (Cambridge Univ. Press, 1993).
11. Bramwell, S. T. & Gingras, M. J. P. Spin ice state in frustrated magnetic pyrochlore materials. Science 294, 1495–1501 (2001).
12. Zhou, Y., Kanoda, K. & Ng, T.-K. Quantum spin liquid states. Rev. Mod. Phys. 89, 025003 (2017).
13. Edwards, S. F. & Anderson, P. W. Theory of spin glasses. J. Phys. F 5, 965–974 (1975).
14. Sherrington, D. & Kirkpatrick, S. Solvable model of a spin-glass. Phys. Rev. Lett. 35, 1792–1796 (1975).
15. Edwards, S. F. & Anderson, P. W. Theory of spin glasses. II. J. Phys. F 6, 1927–1937 (1976).
16. Binder, K. & Young, A. P. Spin glasses: experimental facts, theoretical concepts, and open questions. Rev. Mod. Phys. 58, 801–976 (1986).
17. Principi, A. & Katsnelson, M. I. Self-induced glassiness and pattern formation in spin systems subject to long-range interactions. Phys. Rev. Lett. 117, 137201 (2016).
18. Principi, A. & Katsnelson, M. I. Stripe glasses in ferromagnetic thin films. Phys. Rev. B 93, 054410 (2016).
19. Kolmus, A., Katsnelson, M. I., Khajetoorians, A. A. & Kappen, H. J. Atom-by-atom construction of attractors in a tunable finite size spin array. New J. Phys. 22, 023038 (2020).
20. Kamber, U. et al. Self-induced spin glass state in elemental and crystalline neodymium. Science 368, eaay6757 (2020).
21. Bak, P. & Lebech, B. Triple-Q-modulated magnetic-structure and critical behavior of neodymium. Phys. Rev. Lett. 40, 800–803 (1978).
22. Forgan, E. M., Gibbons, E. P., McEwen, K. A. & Fort, D. Observation of a quadruple-Q magnetic-structure in neodymium. Phys. Rev. Lett. 62, 470–473 (1989).
23. McEwen, K. A. & Zochowski, S. W. Magnetic phase-transitions in neodymium. J. Magn. Magn. Mater. 90–91, 94–98 (1990).
24. Zochowski, S. W., McEwen, K. A. & Fawcett, E. Magnetic phase-diagrams of neodymium. J. Phys. Condens. Matter 3, 8079–8094 (1991).
25. Forgan, E. M. et al. Field effects on the antiferromagnetic ordering of neodymium. J. Magn. Magn. Mater. 104, 911–912 (1992).
26. Lebech, B., Wolny, J. & Moon, R. M. Magnetic phase-transitions in double hexagonal close-packed neodymium metal commensurate in 2 dimensions. J. Phys. Condens. Matter 6, 5201–5222 (1994).
27. Moon, R. M., Koehler, W. C. & Cable, W. Magnetic structure of neodymium. J. Appl. Phys. 35, 1041–1042 (1964).
28. Everitt, B. A. et al. Helimagnetic structures in epitaxial Nd/Y superlattices and alloys. Phys. Rev. B 56, 5452–5460 (1997).
29. Goff, J. P., Bryn-Jacobsen, C., McMorrow, D. F., Ward, R. C. C. & Wells, M. R. Formation of uniform magnetic structures and epilayers in Nd/Pr superlattices. Phys. Rev. B 55, 12537–12545 (1997).
30. Bagrov, A. A., Iakovlev, I. A., Illasov, A. A., Katsnelson, M. I. & Mazurenko, V. V. Multiscale structural complexity of natural patterns. Proc. Natl Acad. Sci. USA 117, 30241–30251 (2020).
31. Eriksson, O., Bergman, A., Bergqvist, L. & Hellsvik, J. Atomic Spin Dynamics: Foundations and Applications (Oxford Univ. Press, 2017).
32. McEwen, K. A., Forgan, E. M., Stanley, H. B., Bouillot, J. & Fort, D. Neutron diffraction study of the magnetic structure of neodymium in a magnetic field. Physica B+C 130, 360–362 (1985).
33. Muldawer, L. & deBergevin, F. Antiferromagnetic-ferromagnetic transformation in FeRh. J. Chem. Phys. 35, 1904–1905 (1961).
34. Green, A. G., Conduit, G. & Krüger, F. Quantum order-by-disorder in strongly correlated metals. Annu. Rev. Condens. Matter Phys. 9, 59–77 (2018).
35. Katsnelson, M. I., Wolf, Y. I. & Koonin, E. V. Towards physical principles of biological evolution. Phys. Sci. 93, 043001 (2018).

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.
© The Author(s) 2022
Methods

Experimental methods. Nd islands were epitaxially grown using the Stranski–Krastanov method on a cleaned W(110) (Supplementary Section 1). The thicknesses of the islands were around 100 ML and represent bulk-like structural, electronic and magnetic properties. The experimental study was performed in a commercial Createc low-temperature scanning tunnelling microscope/atomic force microscope, operating at a base temperature of 5 K. The temperature-dependent study was done by means of a Zener diode attached to the SPM head, allowing us to reach stable conditions between 5 K and 20 K. To obtain spin contrast we used both Cr bulk tips and Nd-coated W tips. They showed similar results, and all data shown in the main text were acquired using Cr tips, while the aging study ( Supplementary Section 2) was performed using Nd-coated W tips. The magnetization images were produced by subtracting majority and minority SP-STM images, and a median filter (1 × 6 pixels) was applied before calculating the FFT. No further data processing was applied to the Q-space images shown. The image data processing was performed using MATLAB.

Simulation methods. The simulations for the static correlation functions were done on a slab of 196 × 196 atoms with a thickness of 16 dhcp layers. The system was described using the same spin model as in ref. 20, that is, with a Heisenberg Hamiltonian using scalar Heisenberg exchange interactions calculated from density functional theory. The simulation protocol consists of first equilibrating the system using 5 × 105 Monte Carlo sweeps and then performing atomistic spin dynamics simulations to obtain a thermal average of the static correlation function S(Q). The atomistic spin dynamics simulation time was set to 100 ps, which was found to be needed to properly capture the glassy dynamics when sampling S(Q). The specific heat data were obtained by taking the numerical derivative with respect to the temperature of the total energy obtained from the same simulations used to calculate S(Q). The simulations were performed using the UppASD 5.0-867 software, and all algorithms used are given in full detail in ref. 36.

Data availability
The data supporting the findings of this study are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Code availability
The codes that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgements
This project received funding from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (grant agreement number 818399). A.A.K. acknowledges the NWO-VIDI project ‘Manipulating the interplay between superconductivity and chiral magnetism at the single atom level’ with project number 680-47-534. This publication is part of the ‘Self-induced spin glasses – a new state of matter’ project (OCENW.KLEIN.493) of the KLEIN research programme, which is (partly) financed by the Dutch Research Council (NWO). B.V. acknowledges funding from the Radboud Excellence fellowship from Radboud University in Nijmegen, the Netherlands. O.E. acknowledges support from the Swedish Research Council, the Foundation for Strategic Research (SSF) and the Swedish Energy Agency (Energimyndigheten). A. Bergman and O.E. acknowledge eSSENCE. A. Bagrov, A. Bergman and O.E. acknowledge financial support from the Knut and Alice Wallenberg Foundation through grant number 2018.0060. M.I.K and O.E. acknowledge the European Research Council via Synergy grant number 854843 (FASTCORR). The numerical simulations were enabled by resources provided by the Swedish National Infrastructure for Computing (SNIC), partially funded by the Swedish Research Council through grant agreement number 2018-05973.

Author contributions
B.V., L. Niggli and U.K. performed the experiments. D.W. and A.A.K. designed and participated in the experiments. B.V. and L. Niggli performed the experimental analysis. A. Bagrov performed the complexity analysis. A. Bergman, D.I., L. Nordström, M.I.K. and O.E. performed and participated in the theoretical calculations. B.V., D.W., A. Bagrov, A. Bergman and A.A.K. wrote the manuscript, while all authors provided input.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-022-01633-9.
Correspondence and requests for materials should be addressed to Alexander A. Khajetoorians.

Peer review information Nature Physics thanks Peter Wahl and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

Reprints and permissions information is available at www.nature.com/reprints.