We demonstrate that a two-dimensional periodic array of spins coupled via RKKY-like exchange can exhibit tunable energy landscapes ranging from robust double-well toward spin glasses. We characterize the magnetic ground states and energy landscapes of such arrays, by the distribution of low energy states, energy barriers, aging relaxation dynamics and the size of the basins of attraction. We identify three phases resulting from singularly varying the RKKY period: a double well phase, a spin glass phase and a multi-well phase. The spin glass behavior results from self-induced glassiness, which is driven by the incommensurability of the RKKY period and the periodic array. We argue that the tunable complexity of these spin arrays can function as an associative memory (Hopfield network) at the atomic scale.

Due to the demand for energy-efficient electronics [1] and pattern recognition [2], there has been recent interest in utilizing brain-inspired concepts in spintronics [3]. A good example is the Hopfield network, a recurrent neural network designed for pattern recognition through associated memory that is based on coupled Ising spin arrays [4]. A given memory pattern is stored as a local energy minimum, or so-called attractor, in a tailored energy landscape comprised of many local minima which can be accessed via stochastic processes. A material realization of the Hopfield model, where the projected magnetization of each spin in the array represents a pixel, requires a coupled spin array based on tunable couplings with a sufficiently
high number of accessible basins of attraction: favorable low energy states separated by finite energy barriers. To this end, the Hopfield network has been intricately linked to the physics of spin glasses [5].

The concept of a spin glass (SG) [6-8], as suggested by Edwards and Anderson in 1975 [9,10], in the thermodynamic limit is characterized by an 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. The classical theoretical SG model [11] assumes fully connected exchange interactions that can be either ferromagnetic or antiferromagnetic. A crucial feature of the glassy behavior is the existence of relaxation processes at all time scales, ranging many orders of magnitude. The slow relaxation processes are particularly spectacular: in a spin glass, any field change causes a very long-lasting relaxation of the magnetization, and the response to an ac field is noticeably delayed. While the energy landscape of an idealized SG such as [11] is too rugged to realize robust basins of attraction due to the infinite distribution of infinitesimally small basins of attraction, understanding and tailoring energy basins with sufficiently large numbers of local minima from simplistic interactions is an important route toward implementing the Hopfield network in material systems. The textbook SG material are the dilute magnetic alloys, like Mn in Cu [10], which have been understood based on magnetic impurities coupled with a long-range indirect exchange interaction with equal probability of ferromagnetic and antiferromagnetic, between randomly distributed moments [11-13]. However, it is still not completely clear, how to describe spin glasses in realistic materials, for example with finite-range interactions, and how atomic-scale ordered arrangements of magnetic ions lead to glassy behavior. Moreover, there is to date no spin-based material system in which the energy landscape, ranging from a deep double well potential toward glassiness landscape can be tuned, varying various parameters.
In this letter, we demonstrate through simulation that a 2D spatially ordered array of interacting spins coupled via a well-defined long-range RKKY interaction [14] can exhibit energy landscapes ranging from robust double-well potentials toward a spin glass, depending solely on the chosen coupling relative to the lattice constant. We show that, depending on the ratio between the periodicity of the RKKY interaction and the lattice constant, that the ground state of the spin lattice is modified ranging from ferromagnetic and antiferromagnetic order, toward more complex order such as checkerboard order. Concomitantly, we show that the distribution of low-energy states changes widely, leading to a large variation in the ground relaxation times depending on one of three identifiable phases. Considering a previously utilized definition of a spin glass [15], we illustrate that the autocorrelation strongly distinguishes the three identified phases, and confirms spin glass behavior for one distinct phase. We finally assess the suitability of the ground states as Hopfield memories, by measuring the size of their basins of attraction and their robustness to perturbations.

We consider simulations of an $8 \times 8$ 2D spin array with an interatomic distance $a$, as shown in Fig. 1a. We consider an exchange Hamiltonian of the form:

$$H = - \sum_{i>j} J_{ij} s_i s_j$$

where $s_i$ represent an Ising spin with the position $i$. The exchange parameter $J_{ij}$ is derived from an isotropic RKKY-like exchange [16]:

$$J_{ij} = \begin{cases} 0, & i = j \\ \frac{1}{r_{ij}^2} \sin \left( \frac{2\pi}{\lambda} n_{ij} \right), & i \neq j \end{cases}$$
where $r_{ij}$ is the distance between positions $i$ and $j$, and $\lambda$ is the period of the RKKY interaction.

In order to investigate the dependence of the spin array properties on the RKKY interaction, we vary the ratio $\alpha = \lambda / a$ (Fig. 1), and analyze the resultant properties of the array. We consider throughout the paper, four values of $\alpha$, namely $\alpha = \{1.375, 2.625, 4.55, 10.5\}$. We use iterative improvement, which is a form of zero temperature spin dynamics that allows single and double spin flips. We initialize a state randomly, run the dynamics and repeat this procedure 2650 times and analyze the distribution of low energy states that are fixed points from the dynamics. We quantify the results in terms of the probability to find the ground state configuration ($P_0$) (Fig. 2c), and the entropy of the distribution of fixed point states ($S$) (Fig. 2c inset). We find three distinguishable phases, which result from the overall interplay between short-range and long-range interactions, with respect to the lattice spacing. The various phases are distinguished by the number of low energy states available in comparison to the energy barriers that separate them, and the relaxation properties, which we discuss later.

For large $\alpha$, $P_0$ is almost 100% indicating that the array easily finds the ground state. We call this the deep double well (DW) phase. In the DW phase, there is a doubly degenerate ground state solution, which are inverses of each other (ferromagnetic in the case shown for $\alpha = 10.5$ in Fig. 2a). Each ground state is a large basin of attraction and separated by a large energy barrier. This can be qualitatively understood resulting from the positive sign of the RKKY interaction for a large range of amplitudes, creating a primarily ferromagnetic interaction. When $4.5 < \alpha < 7$, the ground state is also ferromagnetic, but there are many other stable low energy states (Fig. 2b) as exemplified for the case where $\alpha = 4.55$, thereby greatly reducing $P_0$. As we later show, the presence of many low-energy states in comparison to the DW case strongly modifies the relaxation dynamics. Therefore, we identify this phase as a multi-well (MW).
When $2 < \alpha < 4$, an interesting case arises as the probability to find the ground state $p_0 \rightarrow 0$. As exemplified for the case where $\alpha = 2.625$, we find that the ground state has a checkerboard-like order illustrating a broken symmetry (Fig. 2a). This is accompanied by a much larger distribution of low-energy states very near the ground state in comparison the aforementioned phases (Fig. 2b). This results from the competition between short-range and long-range interactions $j_{ij}$ of both signs, giving rise to frustration inducing a spin-glass like landscape, similar to magnetic domains previously described by the concept of a “self-induced” glassiness [17-19]. These states have relatively small basins of attraction and are separated by low energy barriers. We identify this as the spin glass (SG) phase, which we characterize in more detail below.

For $\alpha < 2$, $p_0$ becomes sensitive to the value of alpha $\alpha$ because of the interplay between the strength of the nearest-neighbor interaction and the lattice spacing. For $\alpha = 1.375$, the nearest-neighbor interaction is four times stronger than all other longer-range interactions, and since the sign is negative the array strongly favors antiferromagnetic order and large basins of attraction. To summarize, by varying $\alpha$, it is possible to tune the spin array into different energy landscapes and we can identify the aforementioned three phases. An alternative illustration of these phases can be seen in the inset of Fig. 2c, where we calculate the entropy $(S(\alpha))$. The value of $S$ roughly traces out $p_0$, illuminating the various phases which we color code for clarity.

Inspired by [15], we perform an aging calculation to further support our claim that a 2D RKKY spin array can be tuned into a SG phase. We consider a randomly initialized array and let this array evolve over time using Metropolis-Hastings algorithm [20] with a temperature $T$ equal to 15 for $t_w$, waiting steps followed by a measurement period in which the aging dynamics are
captured by calculating the autocorrelation function between the state at $t = t_w$ and a later time $t$ as the system continues to evolve using Metropolis-Hastings algorithm with temperature $T = 15$:

$$C(t_w + t, t) = \frac{1}{N} \sum_i s_i(t_w) \cdot s_i(t_w + t)$$

where $N$ is the number of spins and, $t, t_w$ are iterations, where each iteration is a flip attempt of either one or two positions on the lattice. $C(t_w + t, t)$ measures the relaxation over time of all states $s_i(t_w)$. Increasing $t_w$ increases the probability that the system has reached a favorable low-energy state, resulting in a longer relaxation time. A value of $C(t_w + t, t) = 1$ for large $t$, indicates that the array has frozen. Since $C(t_w + t, t)$ depends on the smoothness or ruggedness of the energy landscape, we can use it to analyze the different phases.

Figure 3 shows $C(t_w + t, t)$ for each phase discussed in Fig. 2, for $\alpha = \{1.375, 2.625, 4.55\}$ and $t_w = \{10, 50, 100, 500, 1000, 5000, 10000\}$. As expected, for the DW phase (Fig. 3a), there is essentially only one relaxation time-scale independent of $t_w$ ($t \approx 10^3$), but the asymptotic value of $C$ depends on the value of $t_w$. These are the characteristics of a ferromagnetic system. The SG phase shows the other extreme. Here, the characteristic relaxation time increases with $t_w$ and the asymptotic value is independent of $t_w$. This illustrates that there exists a rich landscape of local minima with multiple spatial scales. The fact that the $C$ does not approach a finite value, regardless of $t_w$, illustrates that the array does not relax to its ground state when starting from a randomized configuration. This confirms that the phase is a spin glass, as was previously seen for the Mn-Cu case [15] (see supplementary materials for aging analysis). The MW phase shows the intermediate case: both the relaxation times and the asymptotic value depend on $t_w$. This suggests that the MW phase contains an energy landscape which combines both
ferromagnetic and spin glass features, ideally suited for associative memory. Moreover, this indicates that the aging analysis is a quantitative measure for distinguishing the various phases.

In order to connect the aging phenomena to the tunability of the RKKY interaction, we quantitatively calculate the resultant energy landscapes depicted in Fig. 1c. Therefore, we consider two properties: (1) the height of the dominant energy barriers, and (2) the size of the basin of attraction of the ground state. To address (1), we calculate a walk through the state space from a ground state to its inverse, considering the minimal energy trajectory for each phase (Fig. 4a). These trajectories were calculated using a restricted random walk algorithm in which the probability of flipping a position is proportional to the exponent of the required energy for the particular flip times a distribution factor. The factor was varied until the algorithm returned a diverse population of trajectories while having a low variance in the maximal energies across the trajectories. From 10000 runs the best trajectory was chosen for each phase. For the DW phase, we find a high-energy barrier and no intermediate local minima, as expected for a double-well potential. Unlike the DW phase, the other phases exhibit a low energy barrier, with multiple local minima as well as plateaus.

In order to address the basins of attraction, we perturb the ground state by randomly flipping $N$ spins, run the zero temperature dynamics and measure the return probability to converge to the starting ground state. In Fig. 4b, we plot the return probability ($\mathcal{R}_0$) for each phase for different $N$ perturbations. The SG ground state has a miniscule basin of attraction under small perturbation, as illuminated by the sensitivity of the return probability. In contrast, the DW ground state is very robust against a large number of perturbations. Only until half the spins (32) are perturbed does the return probability diminish to 50%, resulting from an equal probability of finding the original
state or the inverse state. The MW has also a quite large basin of attraction, similar to the DW. For the functionality of the Hopfield network, the MW is the most interesting because it combines a relatively large number of local minima that have sizable basins of attraction. To be more precise, for the investigated 8 x 8 lattice, we obtain between six to twenty memory states, depending on our definition of a suitable basin of attraction. The maximum memory capacity of a Hopfield network for an 8 x 8 lattice of spins is eight states. We also note that our conclusions are not determined by the geometry of the lattice, and increasing the number of spins in the lattice introduces more complexity [20].

In conclusion, we demonstrate that a well-ordered array of spins coupled with a defined long-range RKKY interaction can be tuned from a double-well potential toward a spin glass state with many local minima, depending on the RKKY modulation relative to the lattice constant. The different phases result from the competing short-range and long-range interactions, which vary in sign and strength, and the interplay with the distance between all neighboring spins. This gives rise to various types of ordered ground states, ranging from ferro/anti-ferromagnetic order, toward more complex states like checkerboard ordering. Heuristically, the emergence of the glassy states is due to the symmetry breaking of the ground state, which is caused by the incommensurability of the RKKY period with respect to the lattice. The results of this illustrates that a tunable energy landscape can be derived from a well-ordered array in 2D, varying one parameter, in contrast to historic studies where magnetic ions were randomly distributed in a bulk metal. It is also interesting that complexity can naturally arise in very simple physical systems with tunable interactions. Moreover, The multi-well potentials found in our simulations can be of a general scientific interest as well; recently such states were discussed in the context of the origin of biological complexity [21]. 2D ordered spin lattices serve as a first step toward creating tailored basins of attraction for a Hopfield model implementation in spintronics systems.
With the advances in spin-polarized scanning tunneling spectroscopy of individual magnetic atoms [22] and the manipulation of long-range oscillatory interactions between magnetic atoms [23-26], it may be possible to realize tailored complex energy landscapes from atoms deposited on top of metallic surfaces.

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Figure 1: (a) A schematic representation of the 2D 8 x 8 Ising lattice coupled via an isotropic RKKY interaction, where the lattice constant is $a$. (b) The considered RKKY interaction landscape between two atoms for three different labeled $\alpha$, considering one of them is placed at the center. The color scale represents the sign of the interaction, and the topography represents the amplitude. (c) A sketch of the energy landscapes corresponding to the various phase, resulting from the various values of $\alpha$, shown in (b). These landscapes correspond to the RKKY periods of their neighbors in (b). We named them: double well (DW), spin glass (SG) multiple (MW).
Figure 2: The characterization of the RKKY lattice as a function of $\alpha$. (a) The ground state at various $\alpha$, where yellow/blue presents spin down/up. (b) The histogram of the energies of low-lying states after 2650 iterations of iterative improvement for each $\alpha$ used in (a) (bin size is 25, the bin width is 25 energy units). (c) The probability of finding a ground state from a random initialization using iterative improvement as a function of $\alpha$. The hues indicate the phase of the lattice; blue is DW, purple is MW, and red is SG. The inset shows the entropy of the distribution over states found by the zero temperature dynamics as function of $\alpha$. 
Figure 3: The calculated aging behavior based on the autocorrelation $C(t_w + t, t)$ for the three phases, as labeled. The aging behavior is shown through waiting times $t_w = \{10, 50, 100, 500, 1000, 5000, 10000\}$. Averaging was performed over 100 randomly initialized runs per $t_w$. Each array was randomly initialized and subsequently ran Metropolis-Hastings at a temperature of 15 for $t_w$ waiting steps, followed by a measurement period in which the aging dynamics $C(t_w + t, t)$ was calculated between the state at $t = t_w$ and a later time $t$ using Metropolis-Hastings with temperature $T = 15$:
Figure 4: The characterization of the energy landscape for each phase, DW (blue), MW (purple) and SG (red). (a) A restricted random-walk algorithm through the state space from a ground state to its inverse, such that the maximum energy of the trajectory is minimal. The walk itself is characterized by the Hamming distance, which is the number of positions in which the state and the inverse differ. (b) The return probability ($R_0$) of the ground state as a function of the initial perturbation ($N = \text{number of spins flipped}$), calculated at zero temperature. For each $N$ the return probability was calculated from 1000 randomly perturbed ground states.