Data mining, dashboard and statistical analysis: a powerful framework for the chemical design of molecular nanomagnets

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Abstract

Two decades of intensive research in lanthanide-based molecular nanomagnets have brought the magnetic memory in molecules from liquid helium to liquid nitrogen temperature. In the pursuit of new derivatives with improved operational temperatures, several “rational” strategies have been proposed and applied through a fluid transfer of knowledge between theoreticians and experimentalists. These have mainly focused on the choice of the magnetic ion and the design of an adequate coordination environment, both in terms of magnetic anisotropy and molecular vibrations. However, much of the progress has been achieved by serendipity, oversimplified theories and chemical intuition. In order to draw conclusions on the chemical design key parameters that govern the physical behavior of molecular nanomagnets in terms of magnetic memory, we apply here a state-of-the-art inferential statistical analysis to a body of over a thousand published experiments. Our analysis shows that the effective barrier derived from an Arrhenius equation displays an excellent correlation with the magnetic memory, and that there are only two promising strategies between all alternatives proposed so far, namely terbium bis-phthalocyaninato sandwiches and dysprosium metallocenes. In addition, we provide an interactive dashboard for visualizing the collected data, which contains all the reported cases.
between 2003 and 2019. This meta-study aims to dispel widespread theoretical misconceptions and will allow researchers in the field to avoid experimental blind alleys.

With the goal of facilitating the chemical design of lanthanide single-ion magnets (SIMs), herein we mined experimental data from over 1400 samples from the first 17 years of SIM research and applied data visualization tools followed by state-of-the-art statistical analysis. This allowed us to obtain a robust data-driven guide on the **key factors governing slow relaxation of the magnetization**. We created a dataset and a dashboard: a freely accessible online interactive app that allows any user to perform qualitative analyses as well as browsing the database. This statistical study represents the first quantitative and data-supported verification/refutation of several widely held "chemical intuitions" in the field of molecular magnetism.

**A brief history of SIMs**

The history of molecular nanomagnets starts at the beginning of the 1990s, when a polynuclear magnetic complex with strong magnetic coupling between *d*-transition ions was reported to display magnetic hysteresis similar to that of hard bulk magnets, but evidencing a quantum tunneling mechanism for the relaxation of the magnetization.\(^1\)\(^2\) This groundbreaking discovery was a great source of motivation for coordination chemists working in molecular magnetism, and the release of an intensified interdisciplinary collaboration with physicists and materials scientists in what nowadays is considered the hottest topic in the field. The collective magnetic behaviour of each of these single-molecule magnets (SMMs) could be approximated to that of an effective anisotropic spin arising from exchange interactions between the spins of each of the individual metal ions. The reversal of this giant anisotropic spin can occur by populating excited spin states and overcoming an energy barrier through a mechanism that was thought to be analogous to chemical reactions. Hence, it was equally described by an Arrhenius equation, an effective potential barrier \((U_{\text{eff}})\) and a pre-exponential factor \((\tau_0)\).\(^3\) Both parameters were not extracted directly from the hysteresis loop, but rather from the combined frequency- and temperature-dependence of the characteristic maxima in the so-called out-of-phase ac susceptibility \((\chi''\))\(^3\)\(^4\). The experimental fact that magnetic hysteresis was observed only at, or near, liquid-helium temperatures was rationalized mainly by a low value for this barrier \(U_{\text{eff}}\); alarmingly little attention was paid to the pre-exponential factor \(\tau_0\) in the Arrhenius equation.\(^5\) Initial models based on effective spin Hamiltonians obtained the relation \(U_{\text{eff}} = D S_z^2\) and concluded that the best strategy to raise \(U_{\text{eff}}\) and therefore improve the hysteresis temperature \((T_{\text{hys}})\) is to maximize the total effective spin \((S)\), rather than the magnetic anisotropy \((D)\), since the latter is a less straightforward target for the synthetic chemist.\(^6\) Despite great effort toward the synthesis of such systems and an abundance of molecules with ever increasing values of \(S\), very little progress was made in the first decade in terms of increasing \(U_{\text{eff}}\) or \(T_{\text{hys}}\).\(^7\)

In the 2000s, a second generation of molecular nanomagnets emerged.\(^8\) This type of single-molecule magnets, commonly known as SIMs, are based on mononuclear complexes containing a single magnetic ion embedded in a coordination environment. These magnetic entities represent the smallest imaginable nanomagnets, with enhanced magnetic anisotropy with respect to the first generation of SMMs, as a result of a strong spin-orbit coupling combined with the
crystal-field interaction with surrounding ligands. The same kind of modelling, based on the Arrhenius equation, resulted in effective energy barriers \( U_{\text{eff}} \) found in SIMs based on rare-earth ions routinely being up to an order of magnitude higher than those of polymeric metal complexes of the \( d \)-block. Same as \( U_{\text{eff}} \), the characteristic maxima in the out-of-phase ac susceptibility \( \chi'' \) moved to higher temperatures, but in contrast the hysteresis temperature \( T_{\text{hys}} \) did not increase significantly in the first years.

Box 1: main physical concepts we employ in the analysis  SIMs are mononuclear metal complexes exhibiting slow relaxation of the magnetization below a certain blocking temperature. Experimentally, this is often manifested in a frequency-dependent out-of-phase ac magnetic susceptibility \( \chi'' \), which may present a maximum above 2K (left), where we define \( T_{B(2K)} \) as the blocking temperature at 10\(^3\) Hz (in presence of a magnetic field). This relaxation dynamics has most often been modelled as an Orbach processes and fitted using the same Arrhenius equation that is employed to describe chemical reaction rates, parameterized by the effective energy barrier \( U_{\text{eff}} \) analogous to the activation energy in chemical reactions, and \( \tau_{o} \), the inverse of the relaxation rate at the high temperature limit. A “full fit” considering other processes in addition to Orbach, such as a direct relaxation, a Raman process or a quantum tunneling of the magnetization, often result in more accurate values \( U_{\text{eff},0} \) \( \tau_{0} \). The best metric for slow relaxation is \( T_{\text{hys}} \), the highest temperature at which the system presents magnetic hysteresis, meaning the highest temperature at which its magnetization depends on the history of the applied magnetic field. Often, the hysteresis curve is “pinched”, signaling a fast relaxation at zero magnetic field.

After the germinal terbium bis-phthalocyaninato sandwich, different chemical families such as polyoxometalates\(^9\) and metalloccenes\(^10\) were synthesized and characterized, also exhibiting slow relaxation of the magnetization of purely molecular origin. The fact that lanthanide SIMs were not restricted to a single chemical strategy, allowing very different kinds of coordination environments, inspired a large community of chemists all over the world to explore many paths in parallel. This has resulted in the reporting of SMM behavior in over 600 complexes and magnetic hysteresis in over 200 complexes, in less than 15 years. A few strategies, derived from seemingly promising results, have been especially prolific. First, phthalocyaninato (Pc) complexes, especially the ones where one of the phthalocyaninato ligands is oxidized and displays an \( S = 1/2 \) radical, have presented good properties\(^11\), inspiring many works centered on Pc and/or radical ligands. Similarly, the introduction of a diamagnetic transition metal ion in the vicinity of the lanthanide was attributed a role in early successes\(^12\) and has been pursued extensively. Later on, the same has happened with metalloccene complexes of type LnCp\(_2\) sandwiches, where Cp = cyclopentadienyl
dianion, which have given rise to a series of world records.\textsuperscript{13–15} However, besides other approaches that have also been paradigmatic (e.g. the use of radicals,\textsuperscript{16,17} and diketonates\textsuperscript{18}), no single chemical strategy has dominated the field in terms of reported examples. Indeed, as we will see below, the vast majority of published SIMs result from molecules that are either a combination of chemically distinct ligands in the same complex or are part of a large variety of individually less frequent ligands.

![Figure 1: Molecular structures of some lanthanide-based SIMs from different representative chemical strategies: a) LnPc \(_2\) \((T_{hys} = 31\) K\textsuperscript{19}, b) LnCp \(_2\) \((T_{hys} = 60\) K\textsuperscript{13}, c) a “mixed ligands” complex \((T_{hys} = 30\) K\textsuperscript{20} and d) effective origin of the “neighbouring diamagnetic TM” family \((T_{hys} = 11\) K\textsuperscript{12}.](image) Indeed, so many studies pursuing independent inspirations have been reported that it is hard for any single scientist to have a proper perspective of what really has been proven to work, despite recent efforts in that direction.\textsuperscript{21–23} Any review of the literature will conclude that both the theoretically-driven design strategies and the free exploration have produced impressive leaps forward, but the fact is that the field risks dedicating valuable time and effort to the exploration of blind alleys due to unverified preconceptions or misunderstandings. This is an opportunity for employing the modern techniques of data analysis and visualization to remedy this knowledge gap. The procedure is labour intensive, since it involves systematically selecting, then reviewing, hundreds of articles and manually extracting from each one the information that is easiest to process, following a consistent protocol. However, after this stage, the data can be analysed with an extensive array of standard statistical and computational tools.
From the point of view of the theory, a common working hypothesis is that one or more of the parameters arising from the theoretical fits of the ac magnetometry, such as the effective barrier $U_{\text{eff}}$ are well correlated with the experimental values for the properties of interest, such as the hysteresis temperature $T_{\text{hys}}$. In other words, a generally unspoken assumption in the field is that there is a simple relation between our theoretical parameterization and the physical properties we want to optimize to make the systems. This however has not been proven, and has actually been challenged in different ways. Over the years, different theoretical approaches have put the focus on the role of different physical processes and therefore different parameters. Thus, here we also aim to evaluate the relation between different physical parameters, and in particular the accord between the fitted parameters and the experimentally determined properties.

Our goal is to produce data-driven guidelines for the chemical design of SIMs, with a secondary goal being understanding the relation between physical variables themselves. We aim to do that by a statistical analysis that quantifies the relationship -or lack thereof- between a series of chemical variables that can be used to describe different lanthanide SIMs and a series of physical variables that have been experimentally determined on the same systems.
Results

An interactive dashboard for lanthanide SIMs

Dashboards are intuitive graphical software applications for data visualization and information management which recently gained widespread popularity thanks to the web-based dashboards to track COVID-19 in real time, starting with the one hosted by the Johns Hopkins University. In this work, we have developed a user-friendly dashboard-style web application named SIMDAVIS (Single Ion Magnet Data VISualization) to host the full set of the most relevant chemical and physical properties of 1405 SIM samples gathered from 453 scientific articles published between 2003 and 2019. SIMDAVIS will allow the chemical community to interact with and visualize the key relationships between chemical structures and physical parameters in SIMs. Our interactive dashboard can be directly invoked by accessing the internet site where it is located. It is organized in 5 tabs: “ScatterPlots”, “BoxPlots”, “Histograms”, “Data” (with 2 subsections: “View Data” & “Download Data”) and “Variables” as we can observe in the tab menu in Fig. 2.

![SIMDAVIS App](image)

Figure 2: Screenshot of the SIMDAVIS dashboard showing a set of selected options offered by the online app to represent the data.

In the SIMDAVIS dashboard, the most versatile source of information is the “ScatterPlots” tab, where the user can represent 9 quantitative physical properties versus one another in a logarithmic scale. It contains a checkbox that allows the user to add a linear regression and it offers the possibility to click on any data point to identify its sample ID, compound and article DOI, facilitating further analysis. In this tab, one can choose between 2 quantitative physical properties to be plotted with respect to each other, as well as a chemical qualitative variable from a dropdown menu, which contains 12 qualitative categorization possibilities. This permits the exploration of hundreds of potential correlations between measured experimental values (such as $T_{axis}$ or $T_{f}$) and the magnetic parameters fitted from physical measurements (such as $U_{eff}$, $U_{eff}$, $f$ or $r_0$), which can be plotted interactively and downloaded as a vectorial PDF file. For example, one can plot the blocking
temperature versus the effective barrier, and every data point will be identified by a color, showing to which chemical family it belongs to. This visual estimate on the relation between descriptors of the magnetic behaviour may uncover trends for specific qualitative variables. For each qualitative variable, each of its categories may be shown or hidden checking their corresponding boxes. For instance, if the categorical variable is of chemical nature, such as the chemical family used to encapsulate the lanthanide, a user may choose to only show SIM samples belonging to the LnPc\textsubscript{2} and the LnCp\textsubscript{2} families.

The next two tabs display the data in complementary ways. The “BoxPlots” tab allows to examine the distribution of each SIMs quantitative property vs a categorization criterion, e.g. we can see the distribution of $U_{\text{eff}}$ values as a function of the elements in the coordination sphere. The boxplot for each category is shown, including the median, the interquartile range (IQR) and the whiskers (1.5xIQR). The “Histogram” tab explores the frequency of different qualitative variables in our dataset. Because of the use of stacked bar graphs the simultaneous analysis of two qualitative variables is available, e.g. we can display, for each chemical family, the number of samples which present magnetic hysteresis. The “Data” tab is a powerful interface to browse the dataset, featuring the possibility to choose the data columns to show, ordering in ascending or descending order and filtering by arbitrary keywords; it also permits downloading all data. Finally, the “Variables” tab gives the user information about the variables contained in the dataset.

Data-driven chemical design of SIMs: what works and what doesn’t?
The goal is now to determine which chemical variables optimize the physical properties. In other words, to perform a main component analysis: what are the main variables the synthetic chemist needs to consider to obtain the desired physical properties? We will first be doing this qualitatively, by analyzing a series of boxplots (see SI section S1)

Let us initially focus on $U_{\text{eff}}$ together with $T_{B3}$, the focus of theoretical and experimental studies respectively. For either parameter, boxplot representations show that the only chemical family with a clearly distinct behavior is the LnPc\textsubscript{2}. The statistical correlation is more important than in the case of the Ln ion, where Dy and Tb are somewhat better than the others, but not as markedly. Similarly, oblate is better than prolate for both properties, whereas non-Kramers ions presents higher $T_{B3}$ but similar $U_{\text{eff}}$ values compared with Kramers ions; here we are in part seeing the influence of the radical phthalocyaninato Tb systems, where the relaxation is slowed down by the spin $\frac{1}{2}$ killing the quantum tunneling but with no significant change in $U_{\text{eff}}$. Other parameters, such as the number or elements of the coordinating donor atoms or the number of coordinated molecules have no significant statistical influence on $U_{\text{eff}}$; however, complexes with 2 ligands and complexes coordinated by nitrogen present consistently higher $T_{B3}$ due to the influence of the LnPc\textsubscript{2} family.

Let us now analyze $T_{\text{hyst}}$, a magnitude that has been much less studied despite being the main justification for this whole field. Here the results change in interesting ways. Here the only ligand family with a distinct positive behavior is the LnCp\textsubscript{2} family, and similarly carbon is the only markedly good element for the coordination sphere. More surprisingly, Erbium has distinctly high hysteresis temperatures, markedly better -on average- than Dy or Tb; this is in sharp contrast with
their relative $T_{B3}$ values, which are consistently much lower in the case of Er. This can also be an indication that searching for equatorial environments, precisely the ones that favour good magnetic properties in Er,$^{26}$ often results in more rigid ligands, and this indicates an underexplored territory. It is certainly possible that certain modifications of [Er(COT)$_2$]$^-$ (or other Er record-bearing complexes) designed to optimize the detrimental effect of molecular vibrations may achieve records that are competitive with DyCp$_2$. Kramers vs non-Kramers is again shown to be irrelevant, but prolate is consistently better than oblate, again in contrast with the opposite behavior which is observed for $T_{B3}$ and $U_{eff}$ and possibly again due to the influence of Er complexes with their more rigid equatorial environments. Finally, both the coordination number and the number of ligands do have an influence on the statistically expected hysteresis temperature, with the best ones being 2 and 7 in the case of the coordination number and just 7 for the number of ligands. As we will discuss below, there are chemical insights to be gained from this.

Further insight is provided by histograms representing the reported presence of magnetic hysteresis, whether full or pinched, as a function of the kind of complex (Fig. 3). Note that we are limited by the minority of the samples where hysteresis or its absence is reported; in the vast majority of the cases this information is lacking. Nevertheless, here it is apparent that certain families such as LnPc$_2$ (and LnCp$_2$) tend to display (pinched) hysteresis.
The “effective barrier”: oversimplified, yet meaningful

A key question is how much the analyses in this field have been affected by the simplified assumption that SIMs relax via an Orbach mechanism. Let’s start by examining the dependency between $\tau_0$ and $U_{\text{eff}}$, the two variables characterizing this process. It has been pointed out that often as $U_{\text{eff}}$ is increased, $\tau_0$ decreases in parallel, leaving $\tau$ essentially constant.\(^5\) We can now check whether the values of $\tau_0$ and $U_{\text{eff}}$ found in the bibliography follow this approximate law indicated for the two-phonon Orbach process in the classical text of Abragam and Bleaney, equation (1).\(^{30}\)
\[ \frac{1}{\tau_0} = C \cdot U_{\text{eff}}^3 \quad \text{with} \quad 10^3 \text{K}^3\text{s}^{-1} < C < 10^5 \text{K}^3\text{s}^{-1} \quad (1) \]

Plotting \( \tau_0 \) vs \( U_{\text{eff}} \) reveals an approximate law \( 1/\tau_0 = C \cdot U_{\text{eff}}^n \) with \( n \) between 2 and 3. Moreover, the data dispersion extends well beyond the expected range, and is skewed towards \( 10 \text{K}^3\text{s}^{-1} < C < 10^5 \text{K}^3\text{s}^{-1} \) (see full analysis in the SI section S5.3). These discrepancies between the experimentally recorded data and the expected equation serve as an independent evaluation of the limitations of a simple Orbach model. The limited (<100) data points in the case of \( \tau_{0,ff} \) and \( U_{\text{eff},ff} \), seem to indicate a somewhat better agreement with equation (1), with \( n = 3, C = 10^2 \). Indeed, \( U_{\text{eff}} \) resulting from an oversimplified model is expected to be less physically meaningful compared with \( U_{\text{eff},ff} \).

Given that \( U_{\text{eff}} \) and the Orbach description seem to be only partially validated in practice, and since we know them to be oversimplifications in theory, a crucial issue that remains is to quantify up to what level the value of \( U_{\text{eff}} \) (or \( \tau_0 \)) are well correlated with the slow relaxation of the magnetization, and whether one needs to employ \( U_{\text{eff},ff} \) instead. Let us proceed in increasing order of complexity. A visual inspection in SIMDAVIS shows that, in the few cases where there is simultaneous information on \( U_{\text{eff}} \) and \( U_{\text{eff},ff} \) their values are very similar (Fig 4a). Furthermore, this partial information is corroborated by the very similar dependencies of \( T_{B3} \) or \( T_{\text{hys}} \) vs either \( U_{\text{eff}} \) or \( U_{\text{eff},ff} \) as well as in the numerical correlations (see SI section S5.1). A categorical analysis (Figs 4b, 4c) shows that the data dispersion is large, meaning it is impossible to predict the experimental behavior for an individual sample merely from its \( U_{\text{eff}} \) value. However, it also demonstrates that a qualitative grouping of samples depending on whether they present a maximum in the out-of-phase susceptibility \( \chi'' \), or hysteresis, pinched or not, has a clear reflection on their \( U_{\text{eff}} \) values. A more thorough numerical analysis (see SI section S6) confirms these trends.

An in-depth statistical analysis of all physical parameters (see SI sections S4, S5, S6) concludes that \( U_{\text{eff}} \) derived from a simple Arrhenius plot is currently the best single predictor for the physical behavior. This means that, whether we are discussing in terms of the presence of maximum in out-of-phase ac susceptibility or the temperature of said maximum, \( U_{\text{eff}} \) is a better predictor than \( \tau_0 \) or, when it appears, \( U_{\text{eff}} \) of a second Orbach process. The number of studies deriving \( U_{\text{eff}} \) from a full fit considering the other physical processes is so low, and the correlation of this “true” \( U_{\text{eff},ff} \) with the “effective” \( U_{\text{eff}} \) is so high, that the data do not support the qualitative observation that \( U_{\text{eff},ff} \) from a full fit is a better predictor for the hysteresis temperature. This does not contradict previous studies which demonstrated that a variation in the Orbach barrier does not fully explain the differences in retention of magnetisation. 21
Figure 4: Main dependencies between the physical variables (see complete analysis in SI sections S5.1 and S6). (a) dependence between $U_{\text{eff}}$ and $U_{\text{eff}, ff}$, (b) distribution of $U_{\text{eff}}$ for samples depending on their qualitative behavior in terms of magnetic hysteresis, (c) distribution of $U_{\text{eff}}$ for samples depending on their qualitative behavior in terms of $T_{B3}$ or, in its absence, of a frequency-dependent out of phase signal $\chi''$. 
Conclusions

We performed a statistical meta-analysis of the first 17 years of the field of lanthanide-based SIMs. We have systematically collected information from over 500 articles and over 1400 samples and built a user-friendly tool for the visualization of all the collected data. Moreover, we carried out an in-depth statistical analysis that allowed grouping the data in clusters based on their chemical and physical properties. From this study, we can highlight two main pieces of information.

In the first place, from the point of view of parametric characterization, the simple Arrhenius fit assuming an Orbach process has been proven to be surprisingly useful. This means that, in general, it is worth it to perform this oversimplified theoretical fit, with the confidence that the effective barrier \( U_{\text{eff}} \) has been proven to present a very good correlation with SMM behavior. Crucially, we have also proven the very different nature of short term magnetic memory in form of the blocking temperature at 100 Hz \( T_{\text{bg}} \) and its long term counterpart in the form of hysteresis temperature \( T_{\text{hys}} \). Strategies that optimize the former are not necessarily best for the latter.

Indeed, in the second place, the chemical roadmap for the preparation of lanthanide coordination complexes with higher \( T_{\text{hys}} \) is now a little more clear: there are, so far, two highly valuable and well-established chemical families, namely bisphthalocyanine terbium complexes and dysprosium metalloccenes. There is therefore value in the optimization within these two families. For example it is now well established that while including a radical in the coordination sphere is not useful by itself, LnPc\(_2\) complexes featuring a radical Pc display enhanced properties.\(^{31}\) Furthermore, the first studies have been made of reduced (divalent) analogues of DyCp\(_2\).\(^{32}\) We find comparatively little value in further pursuing chemical strategies that have been amply explored and never yielded hysteresis above 10 K. On the other hand, we also evidence that there is, of course, value in chemical ingenuity and exploration, in the quest for a third useful family, which according to our results might well be based on equatorial erbium complexes, since these display consistently high \( T_{\text{hys}} \) values. Note that a few complexes included in our data fall into ill-defined families such as “Mixed ligands” or “Other families”, and yet present excellent hysteresis temperatures. It is entirely possible that the next family of record-setters is related to one of promising candidates such as

\[
\text{[Ln(THF)]_5}^3+ \quad (L = \text{BuPO(NHPr)}_2)\]  \(^{20}\) (Fig. 1c), \[
\text{[Dy(Cy_3PO)(H_2O)]_3}^{13+} \quad (\text{Cy}_3\text{PO} = \text{tricyclohexylphosphine oxide})\]  \(^{33}\) \[
\text{Dy}_4(\text{bzhdep}-2\text{H})(\text{H}_2\text{O})_4(\text{NO}_3)_4 \quad (\text{bzhdep} = \text{pyrazine-2,5-diyl-bis(ethan-1-yl-1-ylidene)-di-(benzohydrazide)})\]  \(^{34}\) \[
\text{[Ln(BIPM}^{\text{TMPS}}\text{)]}^3+ \quad (\text{BIPM}^{\text{TMPS}} = \{\text{C(PPh}_2\text{NSiMe}_3\text{)}_2\}_{2}\}^{35}\) \[
\text{Dy(bbpen)}X \quad (X = \text{Cl or Br}) \quad \text{H}_2\text{bbpen} = \text{N,N'}-\text{bis}(2\text{-hydroxybenzyl})-\text{N,N'}-\text{bis}(2\text{-methylpyridyl})\text{ethylenediamine})\]  \(^{36}\) \[
(\text{NN}^{\text{TMPS}}\text{DyI(THF)})_2 \quad (\text{NN}^{\text{TMPS}} = \text{fc(NHSi'BuMe}_2\text{)}_2, \text{fc} = 1,1'-\text{ferrocenediyl}, \text{37}) \quad \text{and} \quad \text{[DyLz}_2\text{(o-vanilin)}]_3^+ \quad (\text{Lz} = 6\text{-pyridin-2-yl-[1,3,5]triazine-2,4-diamine})\]  \(^{38}\) (Fig. 5). For example, two axial phosphine oxide ligands with very bulky substituents seem to function in a similar way as LnCp\(_2\) despite the five equatorial H\(_2\)O molecules. This strategy does not seem to be restricted to phosphine oxides and deserves to be explored further: as we have seen, complexes with 7 ligands have median values of \( T_{\text{hys}} \) close to 10 K, as high as those with 2 ligands.
At the same time, here we offer SIMDAVIS, a dashboard that allows interactive navigation of SIM data. This kind of tool was utterly missing in the field of molecular nanomagnets and paves the way for further studies much beyond the current work. Perhaps more importantly in the wider perspective of design of new materials\textsuperscript{39} and new molecules,\textsuperscript{40} the data mined in this work may serve in the future as annotated training data set for the development of new web scraping systems to retrieve chemical data\textsuperscript{41,42} or even word embeddings\textsuperscript{43} from the scientific literature.

\textbf{Figure 5:} Promising systems for development of new high-$T_{\text{hys}}$ SIMs, chemically distinct from each other and from the TbPc$_2$ and DyCp$_2$ categories.\textsuperscript{33–38} See also Fig. 1c.\textsuperscript{20} (Color scheme for atoms: green, P; cyan, Dy; gray, C; blue, N; yellow, Si; orange, Fe; red, O; magenta, Cl or Br; purple, I. Hydrogen atoms are not shown for clarity.)
Methods

Data gathering. The process started by the collection and organisation of data. We employed the following search criterion for the manuscript: the articles are searched via Web of Science, employing this code:

\[ \text{TOPIC: TS=([lanthan* OR 4$f$ OR "rare$earth") AND ((single NEAR/1 magnet*) OR "slow relaxation")}} \]

Timespan: 2003-2019

Similarly, we employed consistent criteria to decide whether an article was included in the study or not: it needed to include certain data on at least one compound with certain requirements, defined as follows. The compound needs to (a) contain one isolated trivalent lanthanide element from the set \( \text{Ln} = \{ \text{Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb}\} \) and (b) contain no other paramagnetic entity, with the only accepted exception being the presence of a single radical in the coordination sphere and (c) present no strong Ln-Ln interaction, in particular meaning the Ln-Ln separation needs to be larger than 5 Angstrom and larger than 3 bridging atoms between neighbouring Ln centres, and there cannot be a radical in the bridge. The data about this compound needs to include at least one of the following experimental determinations:

(a) \( \chi'' \) vs \( T \) with at least one frequency \( f \) in the window \( 0.9 \text{ kHz} \leq f \leq 1.1 \text{kHz} \) and at a field \( B \) in the window \( 0 \leq B \leq 2 \text{T} \) and/or

(b) \( U_{\text{eff}} \) and/or

(c) \( T_{\text{relax}} \) at sweep speeds \( v \) in the window \( 0.05 \text{T/s} \leq v \leq 0.3 \text{T/s} \).

Further details including the classification in chemical families and the criteria for data extraction are provided in SI.

Shiny App. The dashboard was programmed employing shiny, an open source R package.\(^{44}\) A Shiny-based GUI suitable for non-R users is available as a dashboard-style web application at \( \text{https://go.uv.es/rosaleny/SIMDAVIS} \). The R packages \texttt{readr},\(^{45}\) \texttt{dplyr},\(^{46}\) \texttt{DT},\(^{47}\) \texttt{ggplot2}\(^{48}\) and \texttt{rcrossref}\(^{49}\) were also employed in the development of the app. This interface allows for parameters in the analysis and subsets of the data to be adjusted and chosen in real time.

Statistical analysis. The statistical analysis was also based on R and included Multiple Correspondence Analysis (Gifi system,\(^{50}\) R homals package,\(^{51}\) details in SI section S4.1), clustering studies (FactoMineR,\(^{52}\) details in SI section S4.2), lognormal modelling (Poisson’s distribution, S4.3), factorial analysis of mixed data (FactoMineR\(^{52}\) and factoextra,\(^{53}\) details in SI section S6) as well as simple linear correlations among pairs of parameters (details in SI section S5.1).

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**Author contributions**

A.G.A, J.J.B. and S.C.S. proposed the SIMs study.

J.C. and Y.D. designed the whole procedure for raw data extraction and classification. Y.D., J.C., A.G.A., J.J.B. and S.C.S. did the manual data-mining. Y.D., J.C., A.G.A., L.E.R. and S.C.S. double-checked the raw data.

L.E.R. and A.G.A. cleaned and organized the raw data into a tidy dataset, conceived and supervised the statistical data analysis.

L.E.R. conceived and programmed the dashboard-style interactive web application for data visualization and analysis.

All authors contributed to the preparation of the manuscript.

**Competing interests**

The authors declare no competing interests.
Annex: articles included in the study

1. Ishikawa, N., Sugita, M., Ishikawa, T., Koshihara, S. & Kaizu, Y. 2003 Lanthanide Double-Decker Complexes Functioning as Magnets at the Single-Molecular Level. Journal of the American Chemical Society 125, 8694–8695. (doi:10.1021/ja029629n)

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