Ultrafast manipulation of magnetism bears great potential for future information technologies. While demagnetization in ferromagnets is governed by the dissipation of angular momentum, materials with multiple spin sublattices, for example antiferromagnets, can allow direct angular momentum transfer between opposing spins, promising faster functionality. In lanthanides, 4f magnetic exchange is mediated indirectly through the conduction electrons (the Ruderman– Kittel–Kasuya–Yosida (RKKY) interaction), and the effect of such conditions on direct spin transfer processes is largely unexplored. Here, we investigate ultrafast magnetization dynamics in 4f antiferromagnets and systematically vary the 4f occupation, thereby altering the magnitude of the RKKY coupling energy. By combining time-resolved soft X-ray diffraction with ab initio calculations, we find that the rate of direct transfer between opposing moments is directly determined by this coupling. Given the high sensitivity of RKKY to the conduction electrons, our results offer a useful approach for fine tuning the speed of magnetic devices.

Lanthanides are increasingly important in technology because their 4f spin moments reach exceptionally large sizes compared to those of 3d transition metals. For applications involving ultrafast spin dynamics, however, the localized nature of 4f magnetism poses an additional challenge compared to its 3d counterpart. Highly confined to the space near their ion, the magnetic 4f electronic states generally are not conduction electrons as in the 3d case, but lie several electronvolts below the Fermi level. Therefore, 4f electrons are typically not directly optically excited. Instead, optical pulses excite the conduction electrons, which mediate the RKKY coupling between the 4f spins. In equilibrium, RKKY acts as a Heisenberg exchange, with a coupling energy expressed as \( J \propto |I|^2 \chi \), in which \( \chi \) is the non-local susceptibility of the conduction electrons and \( I \) is the on-site exchange integral between the 4f states and the conduction electrons. The participation of dispersive electronic states renders RKKY exceptionally sensitive to external factors. For example, the oscillating nature of \( \chi \) is central for the large variation in magnetic ground states in the lanthanide metals. \( \chi \) also promises new routes towards ultrafast control of the RKKY coupling between 4f moments, such as by tuning the electronic occupation near the Fermi level. Similarly, the strength of the RKKY interaction depends strongly on the on-site exchange integral \( I \), which is determined by the orbital overlap of the 4f and conduction electron clouds, and therefore strongly depends on the 4f occupation.

This warrants a systematic investigation into the role of 4f occupation on ultrafast magnetization dynamics. Previous attempts to address this question have focussed on ferromagnetic (FM) lanthanide metals, which limits the comparison to three heavier lanthanides (Gd, Tb and Dy) and rules out demagnetization channels that do not involve interactions with the crystal lattice, such as the transfer of angular momentum between antiparallel spins. While reports of ultrafast 4f spin dynamics in antiferromagnets are scarce, one experiment performed on an antiferromagnetic (AF) lanthanide suggested the existence of this channel, which has been proposed as a route to overcome speed bottlenecks associated with the lattice. However, a systematic study of elemental lanthanide metals is hindered by the large variety of different crystal structures and magnetic phases they exhibit, such as spin helices and spin spirals, further complicating a meaningful comparison.

Here, we facilitate a direct comparison of 4f dynamics under comparable conditions by studying ultrafast magnetization dynamics in a series of lanthanide intermetallic antiferromagnets with nearly identical crystal and magnetic structures all across the lanthanide series. This approach allows us to single out the influence of 4f occupation. While demagnetization timescales are found to differ by nearly two orders of magnitude between materials, the corresponding angular momentum transfer rates clearly exhibit a scaling relation known as de Gennes scaling. Our ab initio calculations identify this as transfer between antiparallel moments and show that it scales with the magnitude of the RKKY coupling between them. Our approach provides a microscopic picture of such AF angular momentum transfer, yielding insight substantially beyond phenomenological models, which often do not consider this transfer channel.

We consider a series of antiferromagnets of the form LnRh2Si2, (Ln is a lanthanide element). These share the same crystal structure and 4f spin arrangement (with \(<2.5\%\) change in lattice parameters; Supplementary Section 3), such that the only appreciable difference between them is the occupation of the Ln ions’ 4f shell. Their collinear, compensated AF arrangement (Fig. 1a) exhibits a mean-field-like temperature dependence (Fig. 1b) and removes the need for considering stray fields and domain effects (Supplementary Section 1.2), which are necessary when considering ferromagnets.
As such, these materials can be regarded as a lattice of AF-ordered Ln ions in a Rh$_2$Si$_2$ cage and can serve as an ideal test bed for comparing dynamics of the 4$f$ moments with varying 4$f$ occupation.

We study this AF order using resonant magnetic soft X-ray diffraction. Exclusive sensitivity to the 4$f$ moments is achieved by tuning the incoming photon energies to the Ln ions’ M$_{4,5}$ resonances ($3d \rightarrow 4f$ excitations). The AF-ordered moment $m$ is extracted from the intensity of the magnetic Bragg reflection (normalized to its saturated value $m_0$; Fig. 2a and Supplementary Section 1). To achieve the high temporal resolution needed for this experiment, we used ultrashort X-ray pulses produced by the femto-slicing facility ‘FemtoSpeX’ at BESSY II (ref. 14).

We excite the materials with 1.55 eV laser pulses, and the response is qualitatively identical in all materials: the excitation suppresses the ordered AF moment in a process that begins with a fast (subpicosecond) drop, followed by a second slower drop (Fig. 2b,c).
The fast drop accounts for a smaller fraction of the total reduction (except for Ln = Sm), and is vanishingly small for the heaviest Ln ions studied (Dy and Ho). However, quantitatively the materials’ response times vary widely, ranging from ~1 ps to over 100 ps.

For systematically comparing the behaviour we observe in the LnRh2Si2 family, the excitation fluence was varied. The total reduction in m scales linearly with fluence up to a material-dependent critical fluence Fc, which also varies widely between materials. We define Fc as the fluence at which the total demagnetization amplitude ∆m reaches m/2. Figures 2d,e present the total demagnetization amplitude m/m0 and the dominant (slower) time constant. The data are presented as functions of normalized fluence F/F0 and the time constants τ are also normalized by τc; their values at the critical fluence Fc (inset), demonstrating similar scaling in all materials, despite the markedly different timescales and 4f filling. Exact τc values are extracted by fitting all data in Fig. 2e for each material (Methods).

Two-step demagnetization is typical for lanthanide systems12. The two timescales are understood as one process, which slows down when thermalization of the electronic and lattice degrees of freedom occurs before demagnetization is complete11,12. Such a case is expected for the large 4f moments of many lanthanides, which require more time to release their angular momentum, compared to the smaller moments in transition metal 3d systems. Nevertheless, different Ln14 ions vary appreciably in their moment sizes μμB (μμB g and J are Bohr’s magneton, the Landé factor and the total 4f angular momentum quantum number, respectively), ranging from 0.7μμB to 10μμB. To account for the varying moment sizes, and given the universal dynamics observed in Fig. 2, we facilitate a more direct comparison of the demagnetizations by considering angular momentum transfer rates α, in units of μμB ps−1 (exact definition in the Methods). These are calculated separately for the two demagnetization steps from the total moment J (Methods), but since they both represent the same physical process, we focus on the slow step (Fig. 3a), which we clearly resolve in all compounds. We find that α exhibits a linear relation to the de Gennes factor G=(g−1)2/2(J+1), which approximates the projection of the spin S on J, squared14. De Gennes scaling has been experimentally demonstrated in several 4f systems15,16 for quantities including the interlayer spin turn angle17,18 and magnetic ordering temperatures19, and therefore also for the strength of RKKY coupling19.

The linear relation we observe strongly suggests that ultrafast demagnetization in LnRh2Si2, antiferromagnets depends on the strength of RKKY coupling between antiferromagnetically aligned moments and is therefore governed by the angular momentum transfer between opposite spins. To test this, ab initio calculations of all primary RKKY couplings were performed. These predict that the interplanar coupling j1 (between antiparallel spins) indeed scales linearly with G (Fig. 3b). By contrast, the in-plane couplings j2 and j3 (Fig. 3c) do not show a clear trend with G (Supplementary Section 7).

The linear scaling in Fig. 3a does not cross the origin. This suggests a contribution from an additional angular momentum transfer channel, independent of G, and therefore independent of 4f occupancy (that is, a process that is nearly the same in all LnRh2Si2 materials). One such process is the dissipation of 4f angular momentum to the lattice through the conduction electrons. To analyse this, one can consider a scenario in which angular momentum transfer between opposing 4f spins is turned off. The 4f demagnetization would then depend on two processes, (1) the transfer of 4f angular momentum to the conduction electrons, and (2) its dissipation from there to the lattice. The first process is governed by on-site exchange (and therefore by G), so one could assume that it is faster than the second process. However, since the conduction electron moment is small, it represents a bottleneck for angular momentum transfer such that process (1) is limited by the rate of process (2), and the observed 4f demagnetization would thus be limited by process (2) in a similar way in all LnRh2Si2 materials. When the 4f spin–spin channel is turned back on, it works in parallel to process (2), so this limit is relaxed by the additional angular momentum transfer rate, leading to the linear trend in Fig. 3a. The angular momentum transfer scenario we describe is sketched in Fig. 3d. The bottleneck aspect is similar to the case of 5d–FM semiconductor20,21. Previous works have discussed another channel primarily in the context of FM systems, in which the 4f shell couples directly to the lattice22. Such a channel should depend on the 4f occupation and on G via the strength of spin–orbit coupling, which shows a non-monotonous dependence on G (ref. 9). While our data confirm that the dominating contribution to the angular momentum transfer rates depends on the strength of the RKKY interaction, we cannot rule out additional contributions within the scatter of the data around the line in Fig. 3a.

Our results underline the importance of angular momentum transfer directly between opposite moments, as a channel that can dominate the entire process. This is in line with reports in other RKKY-mediated systems, such as the AF phases of lanthanide metals. Notably, in metallic Dy, which harbours FM and AF phases in different temperature ranges, an efficient demagnetization channel in the AF phase was recently observed, which is absent in the FM phase2. This is understood as the RKKY-mediated spin–spin channel we discuss here, and these observations are also in line with 4f demagnetization in AF metallic Ho2. However, demagnetization in the FM systems Tb and Gd reportedly also exhibited an ultrafast channel1 like AF Dy. The authors of the Dy work2 concluded that

![De Gennes scaling and RKKY coupling](image-url)
this was an extrinsic effect due to spin transport. The differences in demagnetization rates between these three isostructural ferromagnets (Gd, Tb and Dy) were therefore understood as due to different coupling strengths between the 4f shell and the lattice, with a particularly weak coupling for the half-filled 4f shell of Gd.

In conclusion, we have investigated the role of direct angular momentum transfer between spin sublattices in the ultrafast magnetization dynamics of 4f anti-ferromagnets. By a systematic comparison of the ultrafast angular momentum transfer rates with ab initio calculations, we find that the rate of this transfer channel is proportional to the magnitude of the anti-ferromagnetic indirect (RKKY) exchange coupling. Our findings open avenues for ultrafast control of magnetization, for example, by tuning indirect exchange coupling through manipulation of the conduction electrons via doping, voltage biasing or applied pressure, or even transiently, for example through photodoping, without affecting the magnitude of the 4f moments themselves. The implications of our results are not limited purely to anti-ferromagnets, as direct angular momentum transfer can also occur between inequivalent spins in, for example, ferrimagnets or alloys such as Gd–Tb, where direct Gd–Tb angular momentum transfer was demonstrated. Such control over angular momentum transfer rates is also essential for the design and optimization of new device functionalities, such as ultrafast all-optical switching, which has been shown to depend on angular momentum transfer between magnetic sublattices. The ability to tune the demagnetization rate of selected sublattices and the transfer rate between them opens the possibility to engineer such devices, either shortening or prolonging the short-lived collective spin states that enable such effects.

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**Methods**

Sample preparation. Samples were single crystals of all seven materials (LnRh$_2$Si$_2$, with Ln = Pr, Nd, Sm, Gd, Tb, Dy, Ho), grown as previously described$^{2}$: Due to the layered crystal structure, the sample surface is precisely perpendicular to the tetragonal [001] axis. The crystals used were approximately 1–2 mm in size, with faces much larger than the pump and probe beam spots. Neél temperatures found in our experiments agree with literature values$^{1}$: for Ln = Pr, Nd, Sm, Gd, Tb, Dy and Ho, $T_N$ was 68 K, 58 K, 64 K, 107 K, 94 K, 52 K and 29 K, respectively.

Resonant X-ray diffraction. All experiments were conducted by fulfilling the Bragg condition for the (001) magnetic reflection using incoming photon energies near the specific, respective dominant M$_1$ for Pr, Nd, Sm and (M$_2d = 4$) and M$_2$ for Gd, Tb, Dy and Ho (M$_2d = 4$). The spectral shapes of the edges are shown in Supplementary Fig. 1. From the width of the (001) reflection around these edges, we estimate effective probe depths of $\sim$4 nm for Ln = Gd and Sm, $\sim$5 nm for Tb and $\sim$7 nm for Pr, Nd, Dy and Ho. All experiments were conducted with a $\sigma$-polarized incoming X-ray light, such that only the $\sigma = \pi$ scattering channel contributes to the magnetic signal. For all materials except GdRh$_2$Si$_2$, the observed dynamics and temperature dependences do not depend on the azimuthal orientation of the sample around the surface normal, so that the ordered moment $m$ in Figs. 1 and 2 is extracted from diffraction intensity as $m \propto \sqrt{I}$ (Supplementary Section 3). The procedure for extracting $m$ from GdRh$_2$Si$_2$ is detailed elsewhere$^{15}$.

Equilibrium resonant X-ray diffraction experiments were conducted at beamline X11MA of the Swiss Light Source$^{32}$ using the RESOXS end station$^{33}$, and at the PM3 beamline at the Helmholtz-Zentrum Berlin. Time-resolved resonant X-ray diffraction experiments were conducted in a ultrahigh vacuum scattering chamber using ultrashort X-ray pulses from the femto-slicing facility at beamline UE56/1-ZPM (ref. 14) at Helmholtz-Zentrum Berlin. The zone plate monochromators used in this experiment provide an energy resolution typically of $\sim$5 eV (Supplementary Fig. 1). Data for GdRh$_2$Si$_2$ is taken from a previously reported experiment$^{15}$.

The pump–probe experimental scheme was conducted at 3 kHz using 1.55 eV (800 nm) $\sigma$-polarized pump pulses. The X-ray repetition rate is 6 kHz such that between every pumped event an unpumped signal is recorded, and no average heating was observed for the presented data. X-ray intensities were collected using between every pumped event an unpumped signal is recorded, and no average heating was observed for the presented data. X-ray intensities were collected using a convolved Gaussian response function of 120 fs width. The temporal resolution is estimated at 120 fs. To account for this, fits to equation (1) were conducted with a convolved Gaussian response function of 120 fs width. The error bars in Fig. 2a–c are taken as $\Delta I = \sqrt{I}$. Reciprocal space scans (as in Fig. 2a) showed no peak broadening or shifting, so only the peak heights were measured in time traces. The X-ray and 1.55 eV pulses arrive nearly collinearly, but the avalanche photodiode does not collect the pump photons, as they are collected in time traces. The X-ray and 1.55 eV pulses arrive nearly collinearly, but the avalanche photodiode does not collect the pump photons, as they are collected in time traces. The X-ray and 1.55 eV pulses arrive nearly collinearly, but the avalanche photodiode does not collect the pump photons, as they are collected in time traces. The X-ray and 1.55 eV pulses arrive nearly collinearly, but the avalanche photodiode does not collect the pump photons, as they are collected in time traces.

Data availability

All datasets contributing to the results in this work are available on an online repository (https://doi.org/10.5281/zenodo.5828162), including data collected upon equilibrium heating (Fig. 1b) and upon photocexcitation.

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Author contributions
L.R. and D.V.V. conceived the project. K.K. and C.K. grew the crystals. Equilibrium resonant X-ray diffraction experiments were performed by Y.W.W., S.-E.L., D.Z., U.S. and L.R., and analysed by Y.W.W. Time-resolved resonant X-ray diffraction experiments were performed by Y.W.W., S.-E.L., D.Z., C.S.-L., N.P. and L.R., and analysed by Y.W.W. First-principles calculations were done by A.E., V.B., D.T. and O.E. Interpretation was done by Y.W.W. and L.R. The manuscript was written by Y.W.W. and L.R. All authors contributed to discussion and revision of the manuscript to its final version.

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