Unconventional Hund Metal in MnSi

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The interplay between electronic itinerancy and localization in magnetic metals rests at the heart of numerous frontiers in condensed matter physics. Fluctuations of magnetic order in metals inevitably couple to charge carriers and are believed to drive phenomena spanning from dramatic violations of Fermi liquid theory to realizations of high temperature superconductivity. The nature of these fluctuations and the paradigms through which they arise are however often debated. In particular, the continued relevance of local electron correlations driven by Hund’s coupling—the electrostatic interaction between electrons in different orbitals on the same atomic site—in metals far removed from insulating states remains an open question. Here we use inelastic neutron scattering to study a canonical example of a weakly magnetic metal known to be far from a localized insulating regime, MnSi, and we demonstrate that correlation effects surprisingly remain essential to forming its magnetic fluctuations. Short wavelength magnons continue to propagate until a mode crossing predicted for strongly interacting quasiparticles is reached, and the local susceptibility peaks at a coherence energy predicted for a Hund metal by dynamical mean-field theory. Our results establish MnSi as a striking example of a correlated Hund metal and showcase a model where scattering between electrons and intertwined orbital and spin fluctuations generates non-Fermi liquid character distant from a Mott instability.
The coupling between itinerant electrons and magnetic order in MnSi has remained of sustained interest for decades. Cubic MnSi fundamentally realizes a metallic ground state with weak ferromagnetic interactions; however its noncentrosymmetric lattice allows a global Dzyaloshinskii–Moriya interaction to create a long period, helical modulation of its ferromagnetically aligned moments. As a result, it has served as a model material at both the long-wavelength limit for exploring topological spin defects (skyrmions) coupling to charge carriers as well as at the short-wavelength limit where weak ferromagnetism provides a rare experimental realization of magnon decay within a particle-hole (Stoner) continuum. Underlying this is the notion of prototypical itinerancy in MnSi’s charge and spin degrees of freedom—a view supported by the observation of strongly screened moments and the success of conventional band-based models in capturing the qualitative features of low-energy magnon propagation.

MnSi nevertheless exhibits surprising hints of significant electronic correlations in experiment. Large electron mass renormalization and strong quasiparticle damping have been detected in both de Haas-van-Alphen and angle-resolved photoemission (ARPES) measurements. An extended non-Fermi liquid (NFL) phase has been identified under pressure, and indications for NFL behavior are observed in the paramagnetic state’s optical conductivity. The root of these departures of a historically model itinerant magnetic system from the expectations of conventional Fermi liquid theory remains an open question.

An effective means of studying the interplay of the charge and spin degrees of freedom in itinerant magnets is via measurements of the dynamic magnetic susceptibility \( \chi''(q,E) \). Inelastic neutron scattering measurements mapping broad regions of momentum \( \mathbf{q} \) and energy \( E \) space directly sample \( \chi''(q,E) \), and, when combined with density functional theory (DFT) paired with dynamical mean-field theory (DMFT) to model the realistic correlation effects, considerable insight can be gained into the interactions underlying the metallic state. For instance, DFT+DMFT modeling can probe the energy scales of local orbital and local spin coherence directly accessible in neutron measurements. This aspect is critical for identifying and understanding the electronic properties of Hund metals, where interactions stemming from local Hund’s exchange \( J_H \) renormalize electronic properties.

In a Hund metal, strong electronic correlations do not stem from the Coulomb repulsion \( U \), between electrons in the same orbital but instead mainly arise from the effects of the interorbital Hund’s exchange \( J_H \) acting within a broad bandwidth orbital manifold—optimally one with an occupation of one additional electron or hole away from half filling (see e.g. Ref. 30 for a review). The separation of spin and orbital screening energy scales is a common feature of these unusual metals, with Fermi-liquid behavior only appearing well below the lowest screening scale. In the separation regime, incoherent phenomenology appears such as bad-metal or non-Fermi liquid behavior. Prominent examples of Hund metals are established in strongly correlated multiorbital compounds such as iron-based superconductors or ruthenates, yet the range of materials this framework applies to remains a subject of active research.
Here we study the high frequency dynamic susceptibility $\chi''(q, E)$ of MnSi and unveil signatures of strong correlation effects consistent with those of a Hund metal. Overdamped low frequency magnon modes disperse within the particle-hole continuum until an anomalous, intermediate energy scale is reached and the modes’ coherence are abruptly lost into a nondispersive continuum of excitations. This energy scale is captured within DFT+DMFT calculations incorporating both $J_H$ and modest on-site Coulomb interactions $U$, where the momentum integrated local orbital susceptibility $\chi''_{\text{loc}}(E)$ peaks at the onset of the magnon continuum. Our results capture the previously reported violations of Fermi liquid behavior in MnSi as seen by optics$^{18}$ and demonstrate that correlations originating from $J_H$ drive substantial renormalization of the electronic spectrum and low energy properties. This establishes MnSi as an example of an unconventional Hund metal, where a lower-lying orbital coherence scale cuts off damped magnon propagation, and suggests a much broader application of the Hund metal framework than previously envisioned$^{29,30}$.

Well-defined spin waves at low energies are known to appear in the ferromagnetic phase of MnSi$^{11}$ however the spin excitations at higher energies relevant for understanding interactions within the metallic state are relatively unexplored. This is largely due to the fact that, as the spin waves disperse upward in energy, they enter the Stoner continuum, access a decay channel into the particle-hole excitations above $E \approx 3$ meV ($q \approx 0.2$ Å$^{-1}$), and rapidly become damped. $^{31}$ Once in the Stoner continuum, these damped magnons continue dispersing toward the zone boundary with a similar spin stiffness as that observed in the low-$q$ regime—an overall phenomenology consistent with a number of itinerant magnets.$^{32,33,34,35}$ Fig. 1 (a) illustrates this progression of magnetic excitations as a function increasing crystal momentum.

Our inelastic neutron scattering measurements begin by exploring spin excitations within the damped regime where the spin waves have already entered the Stoner continuum. At these $E$ and $q$ values, the low-energy/long-wavelength helical modulation of the ferromagnetic state can be neglected, and the system may be viewed as damped ferromagnetic spin waves. Figs. 1 (b-e) illustrate the dispersion of these excitations about the $q$=(-2, 0, -1) magnetic zone center both upon entering the Stoner continuum above 3 meV and their transformation into a continuum of non-dispersive fluctuations at higher energies. Notably, this high energy continuum develops before the magnetic zone boundary is reached at $\xi=0.5$ r.l.u., which is further illustrated in Fig. 2.

Figs. 2 (a-b) show the parameterization of the spin fluctuations as they disperse within the Stoner continuum of MnSi. Excitations along high symmetry directions are shown as they broaden in $q$, and the modes’ lifetimes decrease. This is parameterized by fitting the excitations to the form $I(E) = \frac{\gamma E}{(E^2-E_0^2) + (\gamma E)^2}$, where $E_0$ is the mode energy and $\gamma$ plotted in Fig. 2(d) parameterizes the damping of the mode. As the damped magnons enter the continuum, $\gamma$ increases linearly with increasing energy, consistent with models of coupled localized spins and conduction electrons$^{36}$. Near 30 meV, the dispersion toward the zone boundary ceases and the values of $\gamma$ diverge. This marks the onset of a true continuum of magnetic fluctuations between $q$=0.6-0.75 Å$^{-1}$ depending on the position within the zone. At all points however, the continuum develops before the wave-vector of the magnetic zone
boundary (marked as dashed lines) in Figs. 2 (a-b). This abrupt decay of spin fluctuations suggests an additional decay channel is opened inside the continuum.

An added decay channel is predicted when quasiparticle interactions are considered. The inclusion of higher-order Landau parameters developed for an interacting ferromagnetic Fermi liquid predicts the stabilization of a gapped mode comprised of transverse spin fluctuations\(^ {37,38} \) (which we label as the \( \Delta \)-mode). While the \( \Delta \)-mode is difficult to measure directly, it can propagate (albeit damped) through the continuum, and when its dispersion crosses with that of the transverse spin fluctuations (shown in supplemental material), an additional damping channel is accessed and the damping rates of the excitations diverge. Model dispersion and lifetimes of these modes in an isotropic model using the \( l=0 \) and \( l=1 \) Landau parameters for MnSi are plotted in Fig. 2 (c). The inclusion of the \( l=1 \) Fermi liquid parameter is the source of the many-body spin orbit interaction in the ferromagnetic phase that stabilizes the \( \Delta \)-mode\(^ {39} \). This feature also implies that the correlations go beyond local correlations since in a local model the \( l=1 \) parameter is zero\(^ {40} \). Despite ignoring the complexity of the multiband nature of MnSi and the naïve breakdown of the small-\( q \) approximations of the theory as the zone boundary is approached, this picture qualitatively captures the onset of the continuum of spin fluctuations in the data. It also hints that strong quasiparticle correlations are essential to understanding the electronic spectrum of MnSi and invites the exploration of local correlation effects in a multiband model.

To understand the local properties, the DFT+DMFT method was employed, where a predominant 3\( d^6 \) character is identified experimentally\(^ {16} \). Figure 3 (a) displays the nonmagnetic band structure from DFT along high-symmetry lines, and, upon the inclusion of moderate local Coulomb interactions of size \( U=2 \) eV and \( J_H=0.65 \) eV on the Mn sites, the multi-sheet Fermi surface is strongly modified. The spectral function \( \Lambda(k, \omega) \) of paramagnetic MnSi (Fig. 3 (b)) acquires substantial band narrowing, sizable shifts of quasiparticle-like dispersion as well as strong incoherence effects away from \( \varepsilon_F \). These features are consistent with strong correlation effects identified in de Haas-van-Alphen and ARPES measurements\(^ {17,15} \), where, for instance, the emerging electron pocket at the M point as well as the strongly renormalized occupied states at the \( \Gamma \) point (both absent in DFT) appear.

The strong correlation effects resulting from moderate interaction strengths for a \( d^6 \) transition-metal compound suggest Hund metal physics as a key driving force. To investigate this further, violations of Fermi-liquid theory—a salient feature of strong Hund metals\(^ {24,26} \)—were sought in the Mn self-energy term \( \Sigma_{Mn} \). By inspecting \( \Sigma_{Mn}(i\omega_n) \) in Matsubara-frequency space and fitting its imaginary part to \( F(\omega_n)=k\omega_n^\alpha \), the exponent \( \alpha \approx 0.5-0.6 \) indeed dramatically deviates from the Fermi-liquid value \( \alpha_{FL} = 1 \) (Fig. 3(d)). NFL character surprisingly appears with the moderate value \( U=2 \) eV, and the \{\( \epsilon_{g}, \epsilon_{d} \)\} orbitals foster stronger correlations than the \( a_{1g} \) orbital. Increasing the Hund’s exchange \( J_H \) not only enhances correlation strength and NFL character, but also increases the orbital-resolved self-energy \{\( \epsilon_{g}, \epsilon_{d} \)\} vs. \( a_{1g} \) differentiation. This role of \( J_H \) as an ‘orbital decoupler’ is a further indication of the relevance of Hund metal type interactions in MnSi\(^ {30} \).
DFT+DMFT calculations yield a Mn(3d) moment 0.3 \( \mu_B \) in agreement with the known saturated moment 0.4 \( \mu_B \).\(^2\) The imaginary part of the local (momentum-integrated) magnetic susceptibility \( \chi''_{\text{loc}}(E) \) can then be analyzed and compared with the experimental results. Figure 4 (a) shows the measured \( \chi''_{\text{loc}}(E) \) with excitations resolved up to 250 meV. A prominent peak appears near 30 meV near the onset of the continuum of spin fluctuations and suggests a low-frequency loss of spin coherence on the local level. In line with this, our calculations unveil intricate features in the frequency-dependent local orbital and local spin susceptibility on the Mn sites, as displayed in Fig. 4 (b). Below 1 eV, the spin susceptibility in Fig. 4 (b) shows first a broad maximum at around 320 meV, whereas the orbital susceptibility first peaks at about twice this value. These energy scales may be associated with the preformation of local moments without yet becoming coherently embedded in the itinerant background. The latter process, surprisingly, takes place at much lower energy scales \( E_\sigma=55 \) meV and \( E_o=38 \) meV for the spin and the orbital degrees of freedom, respectively. The splitting of these two coherence scales in energy is a key feature of Hund metals\(^29\).

The observation of these coherence scales well below 100 meV with a relatively modest \( U \) and the loss of coherence \( E_o\leq E_\sigma \) is unusual. The prominent role of orbital fluctuations is likely endemic to MnSi’s crystal structure that provides a trigonal setting in an overall cubic symmetry, and within this unique environment, strong orbital fluctuations manifest that only become coherent at a comparatively low energy. Figure 4 shows, both for experiment and theory, that well below these coherence scales, the local susceptibilities are linear in frequency, in accordance with finally reaching a Fermi-liquid state\(^29\). Note that while the local susceptibilities are computed for fixed \( T=300 \) K, they still adequately represent the energy spectrum down to the Stoner-continuum onset, and experimentally, the temperature dependence in this energy range is very weak (see supplemental material).

The loss of orbital coherence seemingly coincides with the point where the dispersion of the damped magnons diverges within the particle-hole continuum. This suggests the breakdown of coherent spin fluctuations is due to the coupling of orbital and spin degrees of freedom as seen in the ferromagnetic Fermi liquid theory\(^37,38\). DFT+DMFT modeling provides microscopic insight into the origin of the spin orbit interactions, and an analysis of the orbital-resolved fluctuations (see supplemental) reveals that the coherence peaks are dominantly of itinerant \( e_g' \) character in the spin channel as well as dominantly of \( e_g-a_{1g} \) and \( e_g'-a_{1g} \) character in the orbital channel. Hence in MnSi, a coupling of spin and orbital fluctuations via the \( e_g' \) states seemingly drives the observed phenomenology. It provides an unconventional example of a Hund metal where first, the orbital coherence scale lies energetically below the spin coherence scale, and second, spin and orbital fluctuations are considerably intertwined. The essential inclusion of \( J_H \) and \( U \) to capture the low-energy properties of MnSi place it within a novel paradigm of unconventional Hund metals. This finding suggests a much broader classification of \( J_H \)-driven correlated metals exists beyond the more widely explored multiorbital iron-based superconductors and ruthenates\(^26,27,28\), and that this new regime persists much further into the weakly-magnetic, highly-itinerant regime than previously envisioned.
Methods

Inelastic neutron scattering experiments.
A single crystal with a mass of 52 grams was used for time-of-flight inelastic neutron scattering experiments at SEQUOIA, Spallation Neutron Source at Oak Ridge National Laboratory. Data were collected with different incident energies $E_i = 18, 30, 70, 150, 300, 700$ meV at $T = 5$ K. Some higher temperature data (up to $T = 300$ K) were also collected at select incident energies; however, no appreciable difference is found for high-energy spin wave excitations even at $T = 300$ K. The background is corrected using the intensity at larger $Q$ zone centers where magnetic contributions are negligible. The magnetic form factor term for Mn$^{3+}$ in the scattering cross section is accounted for and removed for after nonmagnetic background contributions were removed. Sample growth and characterization details can be found in Ref. 42. Data were analyzed using the software package Horace.  

Ferromagnetic Fermi Liquid calculations.
We use the version of the Ferromagnetic Fermi liquid Theory (FFLT) where the Fermi Liquid phenomenology is augmented with the Ginzburg-Landau theory for a small moment ferromagnet. The transport and dynamical equations for the FFLT follow from the steady state and time evolution equations of the magnetization distribution function. This leads directly to the prediction of the existence of the amplitude mode when the $l = 1$ Fermi liquid parameter is included we used other experimental measurements to get the magnitude of the amplitude mode for MnSi. This is predicted to be about 50 meV at $q = 0$ and it drops to about 25 meV when the Goldstone mode crosses with the amplitude mode inside the particle hole continuum. The spin hydrodynamic equations were developed here to make the plot of Fig 2(c) as detailed in the supplementary material.

DFT+DMFT calculations.
The cubic B20 crystal structure (space group P2$_1$3) of MnSi has an internal trigonal distortion along the <111> cube diagonal which breaks inversion symmetry. As a result, there are seven Si neighbors for each Mn, grouped into three classes of different bond lengths (see supplemental material). The primitive cell includes four formula units, with all Mn(Si) sites being symmetry equivalent, and the Mn(3d) states split into three symmetry classes: $e_g$ (degeneracy 2), $a_{1g}$ (degeneracy 2) and $a_{1g}$ (degeneracy 1). A charge self-consistent combination of density functional theory (DFT) and dynamical mean-field theory (DMFT) is employed for the one-particle properties. A mixed-basis pseudopotential method is used for the DFT part with the generalized-gradient approximation. Within the mixed basis, localized functions for Mn(3d), Si(3s), (3p) and Si(3d) are utilized to reduce the plane-wave energy cutoff. The correlated subspace for the DMFT part consists of the effective Mn(3d) Wannier-like functions as obtained from a projected-local-orbital formalism. A five-orbital Slater-Kanamori Hubbard Hamiltonian parametrized by a Hubbard $U$ and a Hund’s exchange $J_H$ is applied on the Mn sites. Four symmetry-equivalent Mn sites are in the primitive MnSi cell with lattice parameter $a$=4.558 Å. The single-site DMFT impurity problem is solved by the continuous-time quantum Monte Carlo scheme in hybridization expansion as implemented in the TRIQS package. A double-counting correction of fully-localized type is used. The system temperature is set to $T = 300$ K. Paramagnetism is assumed in all the computations and spin-orbit coupling is neglected. One-particle spectral information is
obtained from analytical continuation via the maximum-entropy method as well as the Pade method.

Experimental estimates of the strength of local Coulomb interactions in MnSi favor a rather low value of $U=2\text{-}3$ eV\textsuperscript{16} which guided the selection in our calculations. For reference, Figure 3(c) shows a comparison of the $k$-integrated spectral function $A_{\text{tot}}(\omega)$ for $U=2$ eV and $U=4$ eV. Because of the large effective Mn bandwidth, there are no essential differences in $A_{\text{tot}}(\omega)$ between these different interaction strengths; however the established renormalization of the strong original Mn(3$d$) peak at -1.9 eV within DFT towards the -1.5 eV observed in valence-band photoemission is better described for the lower $U=2$ eV value. The site and orbital projection $A_{\text{proj}}(\omega)$ unravels the strong intermixing between especially Si(3$p$) and Mn(3$d$) states and shows that electronic correlations are effective in shifting the Si states into lower-energy regions. The remaining contributions close to $E_F$ within the Mn manifold demonstrate that the Mn-$e_g$ orbital supports the itinerant-electron character strongest.

DFT+DMFT calculations yield the moment value of 0.3 $\mu_B$ at 300 K which differs from the larger reported local moment of 2.2 $\mu_B$ obtained via Curie-Weiss analysis of the high temperature susceptibility\textsuperscript{53,54}. This supports the arguments of Moriya and Kawabata\textsuperscript{55}, who demonstrated that the true local-moment response can be obscured via long-range spin fluctuations near the zone center producing a tail in the magnetic susceptibility well above $T_c$. As $q$-dependent spin-fluctuations are not included in single-site DMFT, the temperature independence of the local magnetic moment holds, and instead our theoretical result describes the screened long-time moment. Computing the instantaneous moment however renders a larger $m_{\text{Mn}}^{\text{in}}=2.73$ $\mu_B$. This strong deviation between long- and short-time moment is another fingerprint of Hund metals\textsuperscript{22}.

Local susceptibilities are computed by DFT+DMFT without charge self-consistency using the Hirsch-Fye quantum Monte Carlo scheme. Analytical continuation of the local susceptibilities is performed with the Stochastic Optimization Method (SOM)\textsuperscript{56} (i.e. Mishchenko method) as implemented in the TRIQS/SOM package\textsuperscript{57}. The standard SOM procedure is combined with projection on a very fine energy grid and subsequent rebinning onto a sparser grid. This approach strongly suppresses the stochastic noise and allows to reliably resolve the low-energy spectral features in the computed susceptibilities.

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Fig 1 | Dispersion of spin fluctuations in MnSi away from the low-q limit. (a) Illustration depicting low-q spin waves entering the Stoner continuum and continuing dispersion toward the zone boundary. A second decay channel is accessed at the crossing with the $\Delta$-mode (dashed line). (b) High energy continuum of spin excitations in MnSi collected at $E_i=150$ meV and $T=5$ K. (c) Low energy dispersion of damped magnons within the Stoner continuum, collected with $E_i=30$ meV at $T=5$ K. (d)-(e) show the dispersion of spin excitations via constant energy slices of $S(q,E)$ at 75 meV and 7.5 meV respectively ($H$ is centered about $H=-2$). Black dashed lines in (b) denote the zone boundaries at $\xi=0.5$ r.l.u. Black dashed circles in (d-e) signal the ring-like spin wave excitation, where the radii are in line with the fitted values from (b-c) and Fig 2.
Fig 2 | Parameterization of high energy spin excitations in MnSi. (a-b) combined spin wave dispersion relation along high symmetry directions, collected with different incident energies. Shade bars in (a) are full-width at half maximum (FWHM) from fits to constant E cuts of the spin wave dispersion, demonstrating the broad magnetic excitation along both E and q directions. Green lines in (b) are fits to the spin wave dispersion in the small q limit, showing the q^2 dependence of spin wave excitations. Dashed lines in (b) indicate the Brillouin zone boundaries. (c) Simulation of the expected lifetime broadening of spin excitations as they disperse within the continuum compared with the experimental data along the <110> direction. (d) damping constants \( \gamma \) from damped harmonic oscillator fits to constant q cuts of spin wave dispersion below 20 meV, collected with \( E_i=30 \) meV at \( T = 5K \). Error bars represent one standard deviation of the data.
Fig 3 | One-particle properties of the MnSi correlated electronic structure. (a) Band structure from nonmagnetic DFT in larger and smaller energy window. Purple color marks the Mn(3d) character. (b) DFT+DMFT $k$-resolved spectral function in paramagnetic phase for $U = 2$ eV, $J_H = 0.65$ eV. (c) $k$-integrated spectral function for different interaction strengths. Top: total spectrum with respective Mn(3d) occupation $n_d$, bottom: orbital-resolved projection onto the Mn(3d) states and the Si states. (d) Imaginary part of the Mn self-energy for different interaction strengths at small Matsubara frequencies $\omega_n = (2n + 1)\pi T$. Left: orbital-averaged $-\text{Im} \Sigma_{\text{Mn}}(i\omega_n)$ (symbols) with fitting function $F(\omega_n) = K\omega_n^\alpha$ (lines). Right: orbital-resolved self energies for $U = 2$ eV and $J_H = 0.35, 0.65$ eV.
Fig 4 | Local susceptibilities for MnSi (a) Local magnetic susceptibility determined via inelastic neutron scattering data plotted as a function of energy. The inset shows the low energy region (below 25 meV) and the linear dependence well below the peak near 30 meV. (b) Local Mn susceptibility calculated via DFT+DMFT calculations for both orbital and spin. Vertical dashed lines mark peaks above which coherence is lost in both the orbital and spin sectors. Error bars represent one standard deviation of the data.

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