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Overdamped magnons in the field-polarized phase of cubic helimagnet

Oleg I. Utesov¹, Jonathan S. White², Priya R. Baral³, Tapas Samanta⁴, Chen Luo⁵, Florin Radu⁵, Luana Caron^{4,5} and Victor Ukleev⁵

Abstract

Skyrmion-hosting cubic B20 alloys are considered a fruitful platform for various spintronic applications due to their pronounced tunability. However, disorder can limit their usability by pinning skyrmions and scattering magnons. Here, we probe the spin dynamics in the field-polarized phase of the skyrmion host $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ using spin wave small-angle neutron scattering (SWSANS). Unlike for typical B20 helimagnets, the corresponding SWSANS data do not possess a crucial cut-off feature but rather show smooth diffuse patterns. Our analytical calculations within the minimal model of a disordered cubic helimagnet reveal unusually strong damping of long-wavelength magnons in contrast with standard collinear spin structures. X-ray circular magnetic dichroism (XMCD) measurements indicate antiferromagnetic coupling between Cr and Mn ions, thus strongly supporting the considered model. Therefore, both experimental and theoretical evidence of the absence of well-defined helimagnon excitations in this material are provided.

Introduction

Noncentrosymmetric magnets with Dzyaloshinskii-Moriya interaction (DMI)^{1,2} are a subject of extensive studies nowadays. They host a family of noncollinear spin textures, including planar and conical helicoids, chiral soliton lattices, isolated skyrmions and skyrmion lattices, merons, hopfions, etc.^{3–14} Importantly, complex non-coplanar magnetic structures, e.g., skyrmions, give rise to nontrivial transport phenomena such as the topological Hall effect^{15,16}. From a practical point of view, promising applications of exotic spin structures in memory devices^{17,18} and unconventional computing^{19–22} are actively discussed. Particularly, periodic patterns of magnetic spirals and skyrmion crystals in chiral magnets are considered as media for magnonic applications^{23–26}.

Cubic B20 alloys, e.g., $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ and $\text{Mn}_{1-x}\text{Fe}_x\text{Ge}$, allow the fine-tuning of their electronic and magnetic properties when x is varied. For instance, it was shown that the spiral pitch essentially depends on x , and the chirality can be switched at some x_c ^{27,28}. Moreover, types and properties of topological textures, including their stability, can be tuned by varying x and dopants^{29,30}. Microscopically, this behavior can be connected with the exchange interaction and DMI variation resulting from the electronic band structure modification with x ³¹. However, in some cases, even relatively small dopant concentrations lead to crucial changes in magnetic properties. In particular, $\text{Mn}_{1-x}\text{Fe}_x\text{Si}$ exhibits only short-range magnetic order at $x > 0.11$, which eventually disappears at $x > 0.17$ ^{32–35}. This property can limit possible practical applications of these materials.

The phase diagram of typical B20 helimagnets (e.g., MnSi) is quite simple: it consists of helical, conical, field-polarized phases and includes a pocket of skyrmion lattice A-phase near the ordering temperature⁸. While the magnetic moments are all aligned with the external magnetic field in the field-polarized phase, this regime is far from trivial. It hosts magnons with a nonreciprocal

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spectrum³⁶

$$\varepsilon_{\mathbf{q}} = A(\mathbf{q} - \mathbf{k}_s)^2 + h - h_{C2}, \quad (1)$$

where A is the spin-wave stiffness, \mathbf{k}_s is the spiral vector oriented along the external magnetic field $\mathbf{h} = g\mu_B\mathbf{H}$ measured in energy units, and $h_{C2} = Ak_s^2$ is the field of the transition between conical and field-polarized phase (see, e.g., Ref. ³⁷). This dispersion law leads, for instance, to the magnetochiral effect for phonons^{38,39}, i.e., a difference in the propagation speed for left and right transverse acoustic phonons.

In the last decade, small-angle inelastic neutron scattering on spin waves with dispersion given by Eq. (1) (henceforth referred to as SWSANS—spin wave small-angle neutron scattering) was established to be a relatively simple yet quantitatively accurate technique to probe magnon dynamics. It allows extracting the spin-wave stiffness and magnon damping parameters^{40,41}. For well-defined magnons, the SWSANS scattering maps roughly correspond to the sum of two circles with the crucial cut-off feature beyond which the signal drastically drops down. The cut-off angle was used for the characterization of B20 helimagnets MnSi ⁴⁰ and Cu_2OSeO_3 ⁴² as well as for the more complex $\text{Co}_8\text{Zn}_8\text{Mn}_4$ cubic alloy⁴¹. Noteworthy, in ref. ⁴¹, it was shown that the magnon damping drastically enhances when the temperature is close to the one of transition to the disordered spin-glass phase⁴³.

In our study, we show that the chiral B20 system $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$, despite being a conventional Bak-Jensen cubic helimagnet⁵ according to its phase diagram^{44,45}, does not possess well-defined magnon excitations in the field-polarized regime. Our SWSANS maps do not reveal any cut-off features characteristic to the earlier studied compounds. For theoretical calculations, we use the model of a cubic helimagnet with a bond disorder, which is strongly supported by our X-ray magnetic circular dichroism (XMCD) measurements showing anti-ferromagnetic coupling between Cr and Mn ions. Our model hints at the possibility of low-energy magnon localization and strong damping of moderate-wavelength magnons, which drastically contrasts with usual collinear magnetic systems. Thus, the remarkable tunability of mixed B20 alloy properties can have an important drawback from the spin dynamics point of view, limiting their prospective applications.

Materials and methods

Sample synthesis

Polycrystalline $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ was synthesized using the arc-melting technique, followed by annealing according to the procedure outlined in Ref. ⁴⁶. Details of the sample preparation are given in Ref. ⁴⁵. The same sample as in the previous elastic SANS study was used.

Neutron scattering

Small-angle neutron scattering measurements were conducted using the SANS-I instrument at the Paul Scherrer Institut (PSI). Neutrons with wavelength $\lambda = 6 \text{ \AA}$ ($k_i \approx 10.5 \text{ nm}^{-1}$) were used. The instrument configuration included both collimation and sample-detector distances of 6 m. Magnetic field was controlled using a 6.8 T horizontal-field cryomagnet (MA7). Background SWSANS signal was measured at the base temperature of 2 K and the magnetic field of 3 T where the spin-wave contribution is suppressed. The corresponding SANS pattern was subsequently subtracted from all the other SWSANS datasets. It allows us to get rid of the diffuse elastic contribution due to impurity scattering (see, e.g., refs. ^{47,48}). Importantly, the resulting SWSANS maps have a signal centered at $\pm \mathbf{k}_s$. The typical acquisition time for each pattern was 2 hours. The analysis of all SANS data was performed using the GRASP software⁴⁹.

X-ray magnetic circular dichroism

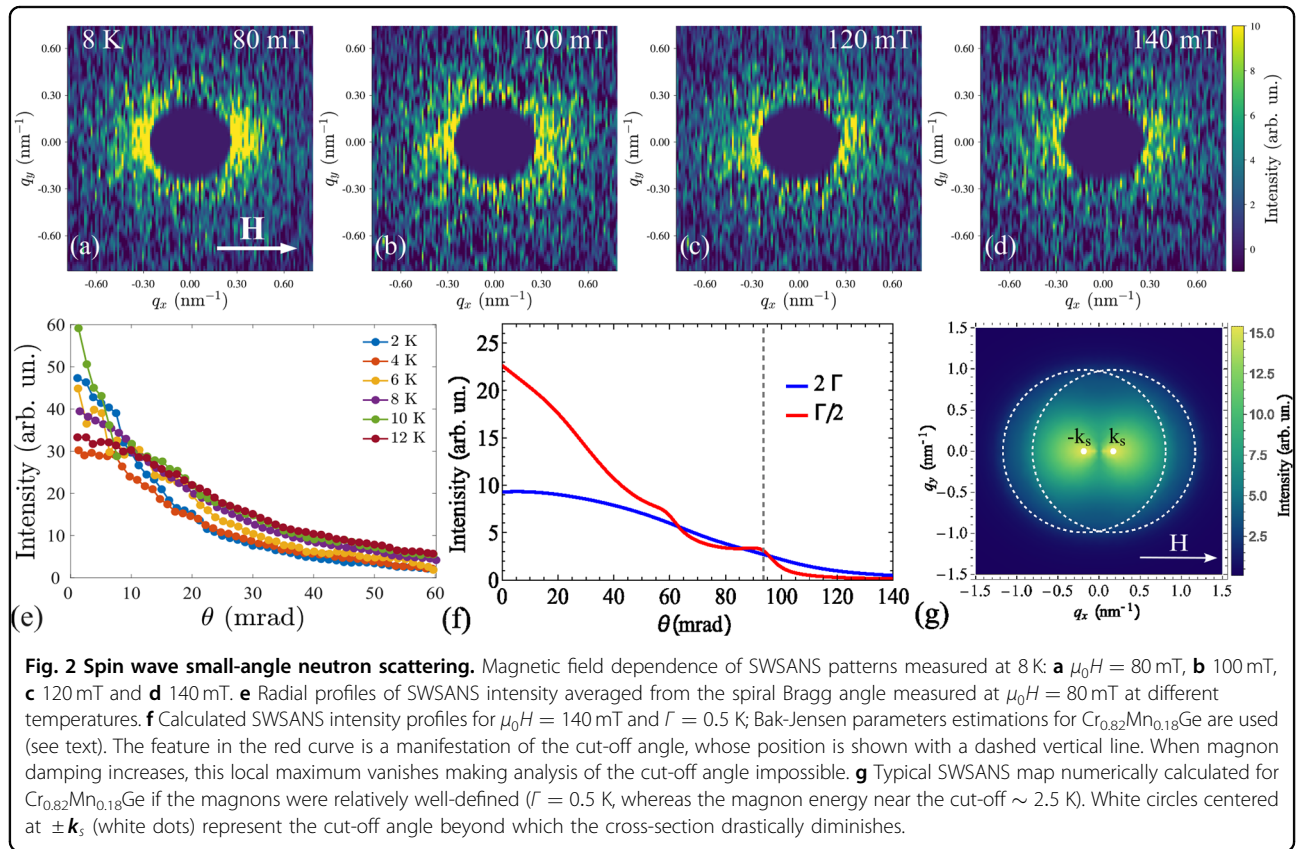
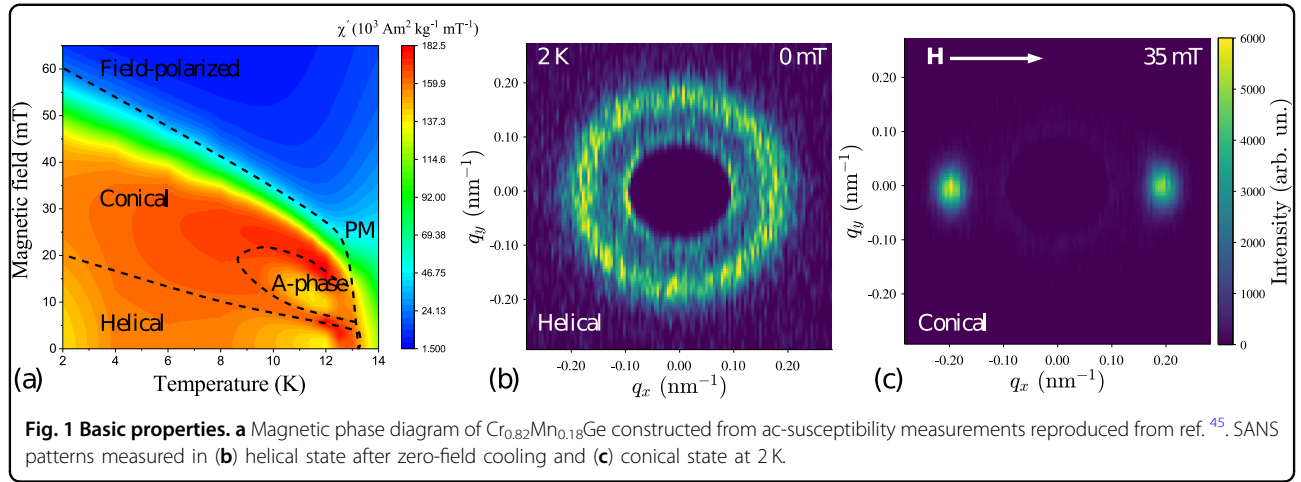
X-ray magnetic circular dichroism (XMCD) measurements were conducted using the VEKMAG instrument at BESSY-II in Berlin, Germany⁵⁰. A polycrystalline ingot of $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ was cleaved in vacuum to prevent surface oxidation. The XMCD measurements were performed in total electron yield (TEY) detection mode, utilizing 77% circularly polarized (C^+) soft X-rays at the Cr and Mn $L_{2,3}$ edges. Magnetic fields ranging from -0.2 T to $+0.2 \text{ T}$ were applied parallel to the x-ray beam.

Results

Spin wave small-angle neutron scattering

According to the previous studies^{44–46} $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ is characterized by a magnetic ordering temperature $T_C \approx 13 \text{ K}$ (Fig. 1a), and a ground state which exhibits a spiral pitch L which varies from $\approx 40 \text{ nm}$ close to T_C to $\approx 35 \text{ nm}$ at $T = 2 \text{ K}$, for which the critical field $\mu_0 H_{C2} \approx 60 \text{ mT}$. Then, using the low-temperature values of the helical (conical) wavevector $k_s \approx 0.18 \text{ nm}^{-1}$ (Fig. 1b and 1c) and $g\mu_B H_{C2} = Ak_s^2$, we arrive at the estimation $A \approx 22 \text{ meV \AA}^2$. Taking the lattice parameter $a \approx 4.8 \text{ \AA}$, we can estimate T_C as Aa^2 , yielding a reasonable value of $\approx 11 \text{ K}$. Also, accounting for the similarity of the phase diagram (Fig. 1a) with that of prototypical B20 helimagnet MnSi ⁸, we conclude that from the macroscopic view, $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ is a typical representative of its class.

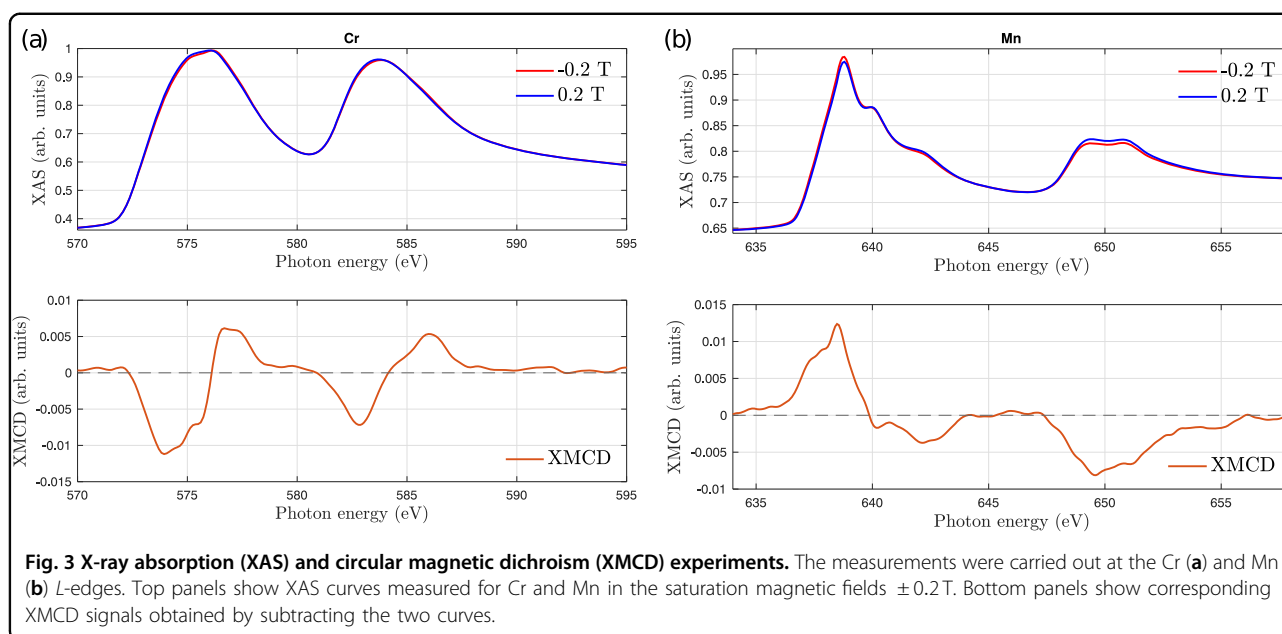
However, the crucial observation of ref. ⁴⁵ is related to the helicoid structures' correlation length, which was shown to be $\sim 100 \text{ nm}$ at relatively low temperatures. It is an apparent manifestation of the short-range magnetic order since the correlation length is only several times larger than the spiral pitch. Moreover, at the same time, it is much smaller than the grain size of the polycrystalline sample $d \sim 1 \mu\text{m}$. Here, the situation resembles recent



studies on the $\text{Mn}_{1-x}\text{Fe}_x\text{Si}$ B20 alloy, where for $x > 0.1$ SANS reveals a finite correlation length for the helioids³⁵, which was associated with strong bond disorder in the form of antiferromagnetic bonds. Remarkably, that observation lies in good agreement with other studies indicating a short-range order phase at these concentrations (see Refs. ^{32–34}).

We proceed with SWSANS studies of $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ at magnetic fields $H > H_{C2} \approx 60$ mT. Typical SWSANS

maps in the field-polarized state of $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ are shown in Fig. 2a–e. The typical acquisition time for each pattern was 2 h. For well-defined magnons with the spectrum described by Eq. (1), one can expect circle-shaped signals with the cut-off corresponding to $q \sim \theta_0 k_i$, where $\theta_0 = \hbar^2 / 2m_n A \sim 0.1$ is estimated for $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ (see Fig. 2fg). However, at all measurement temperatures from 2 K to 12 K, we observed only weak diffuse scattering without any cut-off feature



(Fig. 2a–e). A similar result was reported for $\text{Co}_8\text{Zn}_8\text{Mn}_4$ in the vicinity of the low-temperature spin-glass phase⁴¹.

Direct comparison of SWSANS scattering maps for $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ and insulating Cu_2OSeO_3 is discussed in Supplementary Note 1 and illustrated in Supplementary Fig. 1. Importantly, for the latter material, the well-defined magnons at the cut-off are characterized by energy ~ 1 K and broadening below the experimental resolution $\Gamma \lesssim 0.05$ K⁵¹. In contrast, for $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ magnons with energies ~ 2.5 K should have $\Gamma \gtrsim 1$ K according to the data analysis, thus being overdamped (see Supplementary Note 2 and Supplementary Fig. 2). Here, this property can be associated with the highly disordered nature of the studied alloy. Noteworthy, typical SWSANS intensity in the field-polarized state is ca. two orders of magnitude weaker than the elastic SANS signals shown in Fig. 1b, c.

X-ray magnetic circular dichroism

Results of the XMCD measurement at $T = 8$ K are shown in Fig. 3. Both Cr (Fig. 3a) and Mn (Fig. 3b) show a finite difference between x-ray absorption (XAS) curves measured at ± 0.2 T, which is well above the saturation field of $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$. The spectral line shape of Cr XAS and XMCD signals is similar to the one of the elemental Cr⁵² with the XMCD changing sign once from negative to positive at both L_3 (570–580 eV) and L_2 (580–590 eV) edges (Fig. 3a). In contrast, Mn spectra show multiplet features (Fig. 3b), indicating more localized orbitals, similar to the case of MnSi ⁵³ and $\text{Co}_8\text{Zn}_8\text{Mn}_4$ ⁵⁴. Strikingly, the sign of the XMCD signal from Mn changes from positive at the main L_3 peak (635–640 eV) to negative at the L_2 edge, which

demonstrates that the net magnetic moment of Mn is anti-aligned to the direction of the magnetic field, and also the magnetic moments of the Cr sub-lattice. The inversion behaviour between L_3 and L_2 edges is generic for XMCD spectra of $3d$ transition metals and does not, by itself, indicate the presence of multiple magnetic environments or phase separation⁵⁵. In our data, the opposite signs of the XMCD at the Cr and Mn edges—as opposed to the well-known sign reversal between the L_3 and L_2 edges of a single element—unambiguously indicate antiferromagnetic coupling between the fully field-polarized Cr and Mn sublattices, consistent with their ferrimagnetic ordering. Hence, $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ is, to our knowledge, the first example of a ferrimagnet among B20 materials. Notably, the antiferromagnetic interaction between Cr and Mn is hinted in the $\text{Cr}_{1-x}\text{Mn}_x\text{Ge}$ phase diagram reported by Sato et al. (see ref. ⁵⁶), showing the spin-glass state for higher ($x > 0.24$) Mn concentrations.

Analytical approach

The model of a helimagnet with bond disorder was introduced in ref. ⁵⁷. In the experimental study³⁵, it was successfully applied to the description of the $\text{Mn}_{1-x}\text{Fe}_x\text{Si}$ behavior main features for $x > 0.1$, where only short-range magnetic order can be observed at low temperatures. In $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$, the spatial inhomogeneity of the exchange interaction is evident from element-selective XMCD measurements in the field-polarized state, which reveal opposite magnetization orientations between Cr and Mn ions (see above). These local antiferromagnetic Cr–Mn bonds compete with the ferromagnetic order of the matrix⁵⁸, leading to a spin-glass state at higher Mn concentrations⁵⁶.

We adopt the main ideas of the bond-disordered model to our current purposes. Our minimal model corresponds to the Bak-Jensen⁵ energy density

$$E = \frac{\alpha + \delta\alpha(\mathbf{r})}{2} (\nabla \mathbf{M})^2 - \gamma \mathbf{M} \cdot \nabla \times \mathbf{M} - \mathbf{M} \cdot \mathbf{H}, \quad (2)$$

where α corresponds to exchange interaction and γ to DMI, we neglect anisotropic contributions and allow for a spatially inhomogeneous exchange parameter. Noteworthy, it is easy to show that other types of disorder (e.g., spatial variations of γ) lead to the same results. For definiteness, we choose the magnetic field $\mathbf{H} \uparrow \hat{z}$ and consider only the field-polarized phase. We use the linearized Landau-Lifshitz equation for the magnetization dynamics, see Supplementary Note 3 for details. It can be conveniently written for the combination of transverse components $m_- = m_x - im_y$ as

$$\begin{aligned} \partial_t m_- &= g\mu_B (i\alpha M \Delta m_- - iH m_- + 2\gamma M \partial_z m_-) \\ &\quad + i\delta A(\mathbf{r}) \Delta m_- + i\partial_j \delta A(\mathbf{r}) \partial_j m_-, \end{aligned} \quad (3)$$

where $\delta A(\mathbf{r}) = g\mu_B \delta\alpha(\mathbf{r})M$. After the Fourier transform in the clean case $\delta\alpha(\mathbf{r}) = 0$, it yields the magnon spectrum of Eq. (1) with $A = g\mu_B \alpha M$ and $k_s = \gamma/\alpha$.

Disorder [two last terms of Eq. (3)] scatters magnons, providing plane wave states with a finite lifetime. To proceed with the calculations, we assume that $\delta A(\mathbf{r})$ is spatially correlated on short distances and has a zero mean value. It is pertinent to note that we could also start with the disorder with a nonzero mean value. On the “mean field” level, it will renormalize energy scales of the problem and hence, e.g., periodicity of the spin structures^{28–30,34,35,57}. In our case, a nonzero $\langle \delta A(\mathbf{r}) \rangle$ results in renormalization of A and k_s in the bare spectrum (1). After subtracting the mean value from $\delta A(\mathbf{r})$, we return to the problem at hand. The choice of a particular correlation function is not important on the qualitative level (until it is a short-range one, see Supplementary Note 4 for details); we take

$$\langle \delta A(\mathbf{r}) \delta A(\mathbf{r}') \rangle = S A^2 e^{-\kappa |\mathbf{r} - \mathbf{r}'|}, \quad (4)$$

where S is a dimensionless disorder strength and κ is its inverse correlation length. The natural assumption is that κ is defined by the lattice parameter scale $1/a$, so $\kappa \gg k_s$.

The first order in S (Born) correction to the magnon spectrum due to disorder-induced terms of Eq. (3) can be obtained using standard methods (see, e.g., ref. ⁵⁹). Explicitly, we have

$$\varepsilon_{\mathbf{q}}' = \varepsilon_{\mathbf{q}} + \Sigma(\omega = \varepsilon_{\mathbf{q}}, \mathbf{q}) \quad (5)$$

where the magnon self-energy due to the scattering on the disorder reads

$$\Sigma(\omega, \mathbf{q}) = \int \frac{d^3 \mathbf{q}'}{(2\pi)^3} \frac{8\pi\kappa S A^2 (\mathbf{q} \cdot \mathbf{q}')^2}{(\kappa^2 + q'^2)^2 (\omega - \varepsilon_{\mathbf{q}'} + i\delta)}. \quad (6)$$

Importantly, the pole for $\omega = \varepsilon_{\mathbf{q}'}$ defines the imaginary part of the self-energy and, thus, the magnon damping, whereas the poles $q' = \pm i\kappa$ correspond to the magnon energy renormalization.

In the general case, the integration in Eq. (6) is quite cumbersome. It can be significantly simplified for low-energy magnons with $|\mathbf{q} - \mathbf{k}_s| \ll k_s$. Using the on-shell approximation $\omega = \varepsilon_{\mathbf{q}}$, we obtain the magnon damping in the simple form:

$$\Gamma_{\mathbf{q}} = \frac{2SA\kappa^4 |\mathbf{q} - \mathbf{k}_s|}{\kappa^3}. \quad (7)$$

In the opposite case of $|\mathbf{q} - \mathbf{k}_s| \gg k_s$, the shift in the magnon dispersion (1) is negligible and we obtain the result for usual ferromagnets

$$\Gamma_{\mathbf{q}} = \frac{2SA\kappa q^5}{3(\kappa^2 + q^2)^2}. \quad (8)$$

If $|\mathbf{q}| \sim k_s$, the damping interpolates between the two limiting cases above, so $\Gamma_{\mathbf{q}} \sim SAq^5/\kappa^3$ but it is essentially anisotropic with respect to $\mathbf{q} = \mathbf{k}_s$ point. This is evident, e.g., from the comparison of equal-energy points $\mathbf{q} = 0$ and $\mathbf{q} = 2\mathbf{k}_s$. For the former, the damping is zero, whereas for the latter $\Gamma \neq 0$. Our findings are illustrated in Fig. 4a, b.

The correction to the magnon energy $\varepsilon_{\mathbf{q}}$ is defined mostly by $q' \sim \kappa \gg k_s$ and hence is regime-independent. For $q \ll \kappa$ it reads

$$\delta\varepsilon_{\mathbf{q}} = -\frac{SAq^2}{3}, \quad (9)$$

so the spin-wave stiffness is diminished due to the disorder.

Now it is evident that low-energy magnons (i.e., those with \mathbf{q} near \mathbf{k}_s) are not “protected” against the disorder. Their kinetic energy is $\propto (\mathbf{q} - \mathbf{k}_s)^2$, whereas the damping $\propto |\mathbf{q} - \mathbf{k}_s|$. Usually, it is a manifestation of the perturbative expansion inapplicability and breakdown of a simple damped plane-wave picture for elementary excitations. In our case, localized magnons with energies inside the gap can emerge in regions of space where δA is predominantly negative. Indeed, in these regions, the local H_{C2} is larger than the average, and the gap is smaller [cf. Equation (1)]. Moreover, upon field decrease, the “condensation” of these magnons should lead to the emergence of local noncollinear order (e.g., conical⁶⁰), similar to the Bose-glass transitions in quantum magnets^{61–64}.

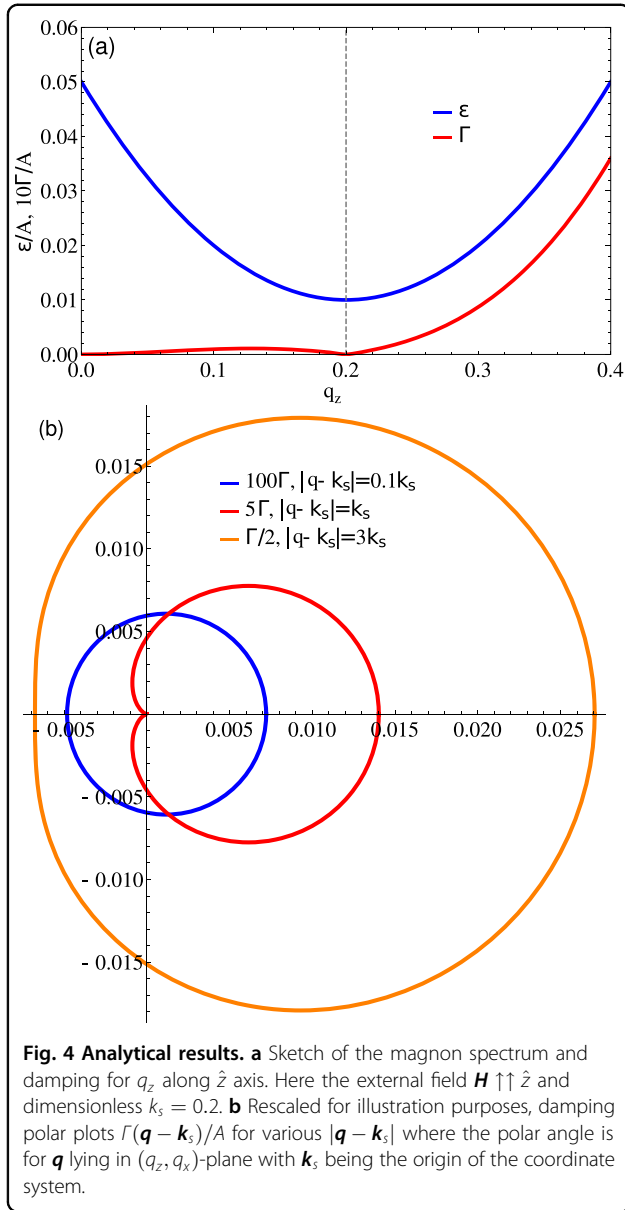


Fig. 4 Analytical results. **a** Sketch of the magnon spectrum and damping for q_z along \hat{z} axis. Here the external field $\mathbf{H} \uparrow \uparrow \hat{z}$ and dimensionless $k_s = 0.2$. **b** Rescaled for illustration purposes, damping polar plots $\Gamma(\mathbf{q} - \mathbf{k}_s)/A$ for various $|\mathbf{q} - \mathbf{k}_s|$ where the polar angle is for \mathbf{q} lying in (q_z, q_x) -plane with \mathbf{k}_s being the origin of the coordinate system.

Notably, the strong damping of low-energy magnons is in contradiction with common knowledge about long-wavelength (i.e., hydrodynamic) modes in ordered 3D magnets with impurities: since the disorder couples to the derivative terms of the corresponding equations of motion, the imaginary part of the spin wave dispersion is parametrically smaller than the real one and the excitations are well-defined. For instance, ferromagnets with fluctuating exchange interaction have $\varepsilon \propto q^2$, whereas the damping scales as $q^{5/9}$, thus being negligible in the limit $q \ll 1/a$ (see also refs. ^{65,66} for a discussion of 3D antiferromagnets).

Turning back to magnons with moderate momenta q , we note that for SWSANS, the most important ones have $q \sim \theta_0 k_i \approx 5k_s$, corresponding to the cut-off angle. For

strong disorder with $S \sim 1$, Eq. (8) yields the damping comparable with their energy ε_q . Hence, these magnons are ill-defined quasi-particles that cannot lead to a meaningful SWSANS signal.

Finally, it is pertinent to note that the polycrystalline nature of the sample should not be important for the conclusions above since $d \sim 1 \mu\text{m}$ is a relatively large length scale. It is easy to show that various orientations of the anisotropy axes and scattering on the grain boundaries provide negligible effects. For instance, the latter yields $\Gamma_q/\varepsilon_q \sim \theta_0^2/k_i d \ll 1$.

Discussion

Using spin wave small-angle neutron scattering and analytical theory for a model of disordered cubic helimagnet, we show that $\text{Cr}_{0.82}\text{Mn}_{0.18}\text{Ge}$ does not possess well-defined magnetic excitations in the field-polarized phase. This conclusion aligns with previous observations of short-range helical structures in this compound.

Our study shows that the pronounced tunability of cubic B20 helimagnets properties can have an important drawback for practical applications when spin structures and phases with ill-defined quasi-particles emerge. In that regime, one can expect that perspective spintronic applications may fall short in terms of their anticipated performance due to suppression of magnon propagation. On the other hand, the disordered nature of these materials can lead to various short-range spin textures, for instance, in regions where the local saturation fields are smaller than the average. One can expect unusual transport properties and responses (see, e.g., Refs. ^{67,68}) under such conditions, which are yet to be studied.

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Author contributions

T.S. and L.C. synthesized and characterized the sample; V.U., J.S.W., and P.R.B. performed neutron scattering measurements; V.U., C.L., and F.R. carried out XMCD experiments; O.I.U. developed the analytical approach; V.U., C.L., O.I.U., and J.S.W. analyzed the data and wrote the manuscript. O.I.U., L.C., and V.U. jointly conceived the project.

Data availability

The data is available from the corresponding authors upon reasonable request.

Competing interests

The authors declare no competing interests.

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