

Disorder-induced damping of spin excitations in Cr-doped BaFe₂As₂

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In doped Hund's metals, such as the iron-based superconductors, effects like charge doping and chemical pressure are often considered the dominant factors. Partial chemical substitution, however, inevitably introduces disorder. Here, we investigate spin excitations in Ba(Fe_{1-x}Cr_x)₂As₂ (CrBFA) by high-resolution resonant inelastic x-ray scattering for samples with $x = 0, 0.035$, and 0.085 . In CrBFA, Cr acts as a hole dopant, but also introduces localized spins that compete with Fe-derived magnetic excitations. We found that the Fe-derived magnetic excitations are softened and damped, becoming overdamped for $x = 0.085$. At this doping level, complementary angle-resolved photoemission spectroscopy measurements show increased electronic localization and a suppression of the nematic d_{xz}/d_{yz} band splitting present in the parent compound. We thus propose a localized spin model that explicitly incorporates substitutional disorder and Cr local moments, successfully reproducing our key observations. Our findings reveal a case where disorder dominates over charge doping in the case of a Hund's metal.

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Introduction. Electronic correlated materials often display multiple competing interactions, each associated with one or more energy scales. Partial chemical substitution tunes interaction energy scales to stabilize a desired ground state. This gives rise to rich composition (x) versus temperature (T) phase diagrams, characteristic of correlated systems [1].

In iron-based superconductors (FeSCs) [2], chemical substitutions often induce the suppression of the antiferromagnetic (AFM) ordered phase and the emergence of high-temperature superconductivity (HTSC) [3–9]. While these effects are typically attributed to charge doping or chemical pressure [10], the role of disorder, an inevitable consequence of substitution, remains unclear. Early scenarios for the physics of FeSCs proposed that disorder could be the driving mechanism behind the observed x versus T phase

diagrams [11,12], but subsequent approaches favored charge doping and chemical pressure effects [13,14].

The parent compound BaFe₂As₂ exhibits a stripe-type AFM transition at $T_N \approx 134$ K, accompanied by a tetragonal-to-orthorhombic structural transition [15,16]. Substitutions suppressing these transitions may or may not induce superconductivity (SC). For instance, Mn substitution (MnBFA) introduces minimal charge doping, mostly affecting the electron pockets [17–19], and some attention was devoted to the effect of disorder acting as the main tuning knob in this phase diagram [20–23]. In contrast, Cr substitution (CrBFA) acts as a hole dopant [24], but the evolution of T_N in CrBFA and MnBFA follows a similar trend as a function of x , with no emergence of SC for both substitutions. Therefore, charge doping alone cannot explain their phase diagrams.

Previous studies have shown that hole doping enhances electronic correlations in BaFe₂As₂ [25,26], an effect often attributed to proximity to a Mott insulating state. However, experimental observations show that the ground state remains metallic even in regimes with localized magnetism. This apparent contradiction can be resolved by the concept of spin-orbital separation in multiorbital systems: The Hund's coupling delays spin screening relative to orbital screening, leading to the so-called Hund's metal behavior. BaFe₂As₂ is

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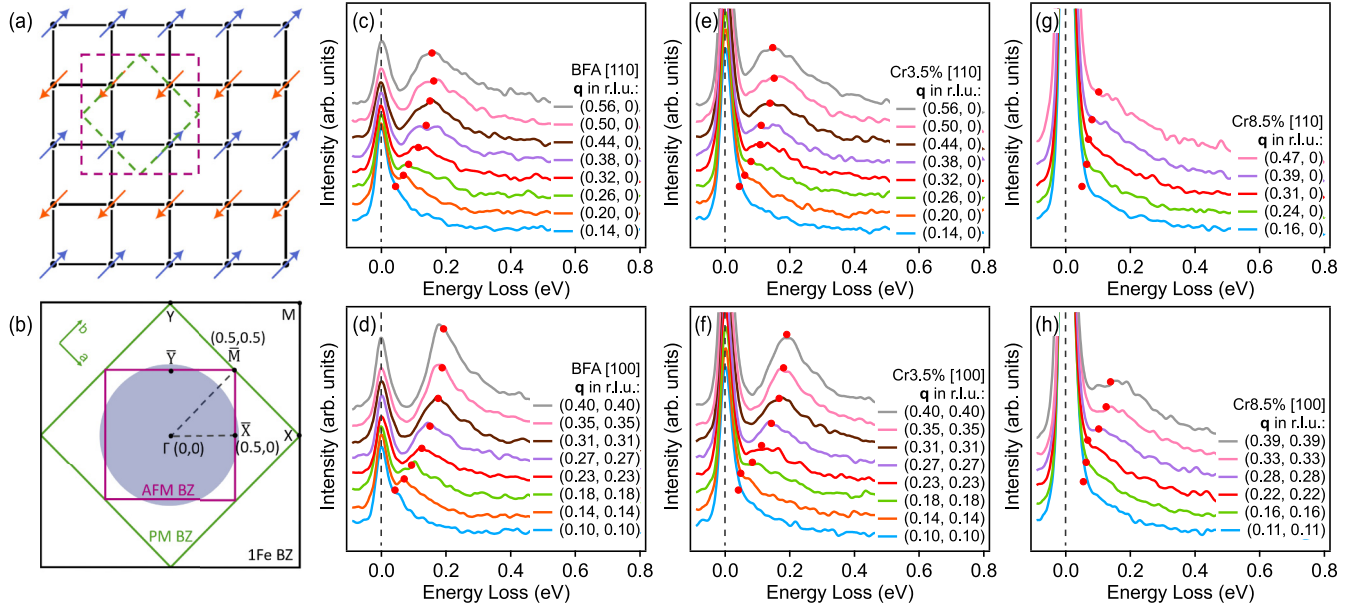


FIG. 1. Overview of RIXS results. (a) Two-dimensional (2D) magnetic lattice with the stripe order. The green and magenta dashed squares show the PM and the AFM phases' unit cells. (b) The respective reciprocal spaces for the 2D first Brillouin zones; the circle indicates the probed $\mathbf{q}$ -range, with the dashed lines showing the high-symmetry directions for the RIXS experiments. The 1Fe BZ (black square) is used for the theory. (c)–(h) Momentum-dependent RIXS spectra for the samples with $x = 0, 0.035$, and 0.085 (BFA, Cr3.5%, and Cr8.5%) and for different directions after background subtraction. The data are shifted for better comparison.

currently recognized as a Hund's metal, where strong Hund's coupling and orbital selectivity govern the degree of electronic correlations, which are expected to increase further with hole doping toward half filling [27–30]. While K substitution on the Ba site preserves itinerancy and superconductivity even at high hole doping, direct substitution at the Fe site, as in the case of Cr, introduces carriers that enhance localization and Fe-sublattice disorder. Resonant inelastic x-ray scattering (RIXS) is proven to be a powerful tool to investigate magnetism with the contrasting itinerant character of electrons and localized character of spins within FeSC phase diagrams [29,31–33].

In this work, we investigate how spin excitations originating from Cr and Fe compete for the ground-state properties across the x versus T phase diagram of CrBFA. To this end, we performed high-resolution RIXS, complemented by angle-resolved photoemission spectroscopy (ARPES) experiments and theoretical modeling. Our combined experimental and theoretical analysis shows that the key features of the data can be understood within a localized spin model, where disorder effects introduced by Cr moments play a central role. This reveals a case of a correlated metal in which disorder dominates over the charge doping effect.

Our results explain why the suppression of the AFM order in CrBFA and MnBFA does not follow the expected trend for charge doping, where Cr nominally introduces twice as many holes as Mn [17,34]. They may also help explain the x – T phase diagrams of Cr/Mn-substituted $\text{CaKFe}_4\text{As}_4$, where the suppression of both HTSC and AFM order similarly deviates from charge doping expectations [35,36]. Altogether, our findings suggest that the absence of SC in these systems is likely linked to the joint effect of increasing correlation and disorder effects on the Fe lattice introduced by Cr and Mn impurities.

Materials and methods. $\text{Ba}(\text{Fe}_{1-x}\text{Cr}_x)_2\text{As}_2$ single crystals, with different concentrations of Cr, were grown using the In-flux method [37]. Sample characterization methods are described in the Supplemental Material [38–41].

The $x = 0, 0.035$, and 0.085 samples (denominated as BFA, Cr3.5%, and Cr8.5%, respectively) were selected for RIXS experiments at the I21 beamline at Diamond Light Source [42], with a detected photon energy resolution of ≈ 30 meV. The energy of the absorption spectra L_3 edge maximum at 709.5 eV was chosen for the momentum-dependent studies. The methods of the sample preparation and data acquisition are detailed in the Supplemental Material [38].

We define the wave vector $\mathbf{q}$ in reciprocal lattice units (r.l.u.) as $(H, K) = (q_x, q_y)a/2\pi$, where $a = 5.598, 5.590$, and 5.616 Å are the in-plane lattice parameters for BFA, Cr3.5%, and Cr8.5%, respectively. These correspond to the magnetic unit cell shown as a magenta dashed square in Fig. 1(a). Since the samples are twinned, we adopt $a = b$ and do not distinguish orthorhombic a and b directions. In this Brillouin zone (BZ), we probe a maximum momentum of $|\mathbf{q}| \approx 0.56$ r.l.u., indicated by the blue circle in Fig. 1(b). High-symmetry points are labeled using the 1Fe BZ (black square) and consistently used for ARPES and theoretical results. ARPES acquisition details are as described in Refs. [17,24] and in the Supplemental Material [38].

To model the magnetic excitations, we employ a minimal antiferromagnetic (AFM) XXZ J_1 – J_2 model on a square lattice, with anisotropy favoring the spins to lie on the S_x – S_y plane. Classically, this system exhibits checkerboard (Néel) order for $J_2 < J_1/2$ and stripe order for $J_2 > J_1/2$. Cr doping (x) is treated as a local disorder in J_2 , the coupling controlling the transition between the ordered phases. At $x = 0$, the system has stripe order; at $x = 1$, it displays Néel order. For

intermediate doping, we randomly replace a fraction x of Fe sites with Cr, assigning J_2 between sites i and j as follows: (1) Fe-Fe, $J_{2,ij} = J_2(x=0)$; (2) Cr-Cr, $J_{2,ij} = J_2(x=1)$; and (3) Fe-Cr, $J_{2,ij} = [J_2(x=1) + J_2(x=0)]/2$. The classical ground state is determined via Monte Carlo simulations and annealing. Excitation spectra are computed by integrating the semiclassical equations of motion: $d\vec{S}_i/dt = \vec{h}_i \times \vec{S}_i$, where $h_i^\alpha = \sum_j J_{ij}^\alpha S_j^\alpha$ is the local exchange field at site i and $\alpha = x, y, z$. Finally, we Fourier transform $\vec{S}_i(t)$ to obtain the dynamical spin structure factor for comparison with experiments. Details are provided in the Supplemental Material and references therein [38,43–59].

Results and discussion. In Fig. 1(c)–(h), we show an overview of our RIXS momentum-dependent spectra for each sample. The probed reciprocal space high-symmetry directions and range are represented by the black dashed lines and blue circle in Fig. 1(b). A background subtraction was performed to better visualize and fit the quasielastic region (which also features low-energy phonon excitations) and magnon excitations. The subtraction method is explained in the Supplemental Material [38,60–62].

The main magnon dispersion is clear even for the Cr8.5% substitution, for which the elastic line is enhanced and the magnon is softened. The magnon appears as a shoulder of the quasielastic peak at low $\mathbf{q}$ and is evident at maximum momentum transfer. To gain more quantitative insight, spectra were fitted to a model composed of an elastic (Gaussian), a phonon (Gaussian), a principal magnon peak, and a secondary inelastic magnonlike peak (Gaussian).

The main magnon peak can be fitted to the imaginary part of the dynamic susceptibility of a damped harmonic oscillator (DHO) ($\chi''(\omega)$), as shown in Eq. (1) [64,65], where the $\mathbf{q}$ dependency reveals the magnon dispersion, I_0 is an intensity constant, γ is the damping coefficient, and ω_0 is the undamped (bare) frequency of the spin excitation as a function of $\mathbf{q}$. The Bose-Einstein distribution is included to reproduce thermal effects:

$$\chi''(\omega) = \frac{1}{1 - e^{-\hbar\omega/k_B T}} \times \frac{I_0 \gamma \omega}{(\omega^2 - \omega_0^2)^2 + 4(\gamma \omega)^2}. \quad (1)$$

We refer to the peak energy of the magnetic excitation as ω_{peak} , i.e., the energy at $\chi''(\omega)$ reaches its maximum.

We note that it is necessary to include the second magnon-like peak in the fitting to accurately describe the shape and sharp width of the main magnon peak, especially for the highest $\mathbf{q}$ values and in the case of the parent compound. The inclusion of this peak is justified by the inspection of Fig. 2(a), wherein we show a direction and polarization survey of our results for the parent compound. As can be observed, a second excitation appears riding on top of the background fluorescence and peaking around ≈ 450 – 550 meV. It is clearer for the [100] (π, π) direction, as indicated by the orange and blue arrows.

Figures 2(b) and 2(c) show examples of fittings for the BFA and Cr3.5% samples. The data after the background subtraction are shown as blue dots, the total fitting as the red line, and the shaded regions as the different fitting contributions, including the elastic line, phonon, main magnon, and secondary magnonlike peak, shown as pink, green, blue, and orange

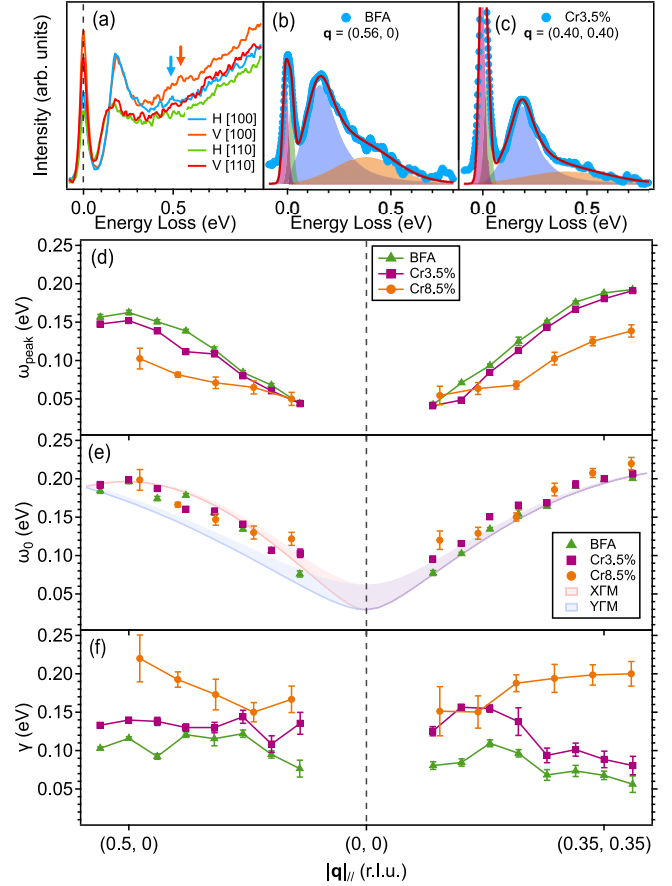


FIG. 2. (a) Polarization-dependent RIXS spectrum of the parent compound (BFA) measured at $|\mathbf{q}| = 0.56$ r.l.u.. (b), (c) Representative fits of the quasielastic and magnon contributions at maximum momentum transfer for the BFA and Cr3.5% samples, respectively. (d) Peak energy of the magnetic excitation ω_{peak} , (e) bare frequency ω_0 , and (f) damping coefficient γ as functions of momentum, for different samples and high-symmetry directions. In panel (e), the solid line shows the linear spin wave dispersion at $L = 0$, using parameters from inelastic neutron scattering fits [63]. The shaded area indicates the dispersion variation with L , considering the 3D Brillouin zone.

areas, respectively. All the fittings for different momentum points for all samples and directions are shown in the Supplemental Material [38]. The need for the secondary magnon peak is particularly evident along the (π, π) direction, where the main magnon peak is sharp but notably asymmetric, which is not expected from the fitted model [65]. At lower $\mathbf{q}$ values and for the doped samples, the secondary magnon peak becomes less pronounced and may even vanish after background subtraction. The dispersive properties of this second peak can be observed for the $\mathbf{q}$ regions where its contribution has relevant weight to the fitting, close to the magnetic zone boundary, as shown in Fig. S11 of the Supplemental Material [38]. This behavior supports the interpretation of the feature as a bimagnon, a bound state of two correlated spin flips on neighboring sites, with well-defined momentum and relatively long lifetime [66–68].

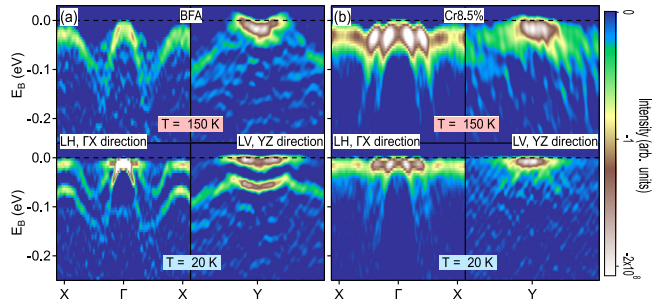


FIG. 3. ARPES band maps second derivatives at paramagnetic and ordered phases for (a) BFA and (b) Cr8.5% samples. The transition temperatures are $T = 134$ K and $T = 79$ K, respectively. The minimum of the second derivatives denotes the Lorentzian-shaped bands' maximum position.

The peak energy of the magnetic excitation ω_{peak} takes into account the finite lifetime of the excitation by including the damping effect. We inspect it as a function of momentum, in Fig. 2(d), where it is possible to observe its energy dispersion as a function of transferred momentum for different dopings and directions. We found that the magnon dispersion is considerably softened for the Cr8.5% sample only, and equally softened in both directions, which contrasts with the Mn8% sample [69].

To further investigate this effect, we can also analyze the trend of the bare frequency ω_0 and the damping coefficient γ , as shown in Figs. 2(e) and 2(f). The ω_0 $\mathbf{q}$ dispersion is nearly sample independent, and we compare it with the results of a linear wave theory, plotted with parameters obtained from inelastic neutron scattering experiments of the parent compound [63]. A good agreement is observed in the probed momentum region. The shadowed area represents the L dependency of the linear wave model, based on a Hamiltonian with a weak out-of-plane interaction component. The X/Y anisotropy of the detwinned sample is evidenced in this model, with our twinned sample dispersion lying in between the curves for the ΓX and ΓY directions.

Along $(\pi, 0)$, γ is nearly $\mathbf{q}$ -independent for the BFA and Cr3.5% samples, and becomes $\mathbf{q}$ -dependent for Cr8.5%, increasing monotonically with $\mathbf{q}$. It suggests shorter magnon lifetimes, likely due to competing Néel-type fluctuations derived from Cr. Along (π, π) , the damping varies nonmonotonically for BFA and, in particular, for Cr3.5%. For Cr8.5%, where the bimagnon peak can be neglected, the damping increases monotonically with $\mathbf{q}$, indicating that a genuine momentum-dependent γ emerges for CrBFA with increasing Cr content. Our results align with studies linking Cr doping in CrBFA to weaker orthorhombicity and more localized (yet still itinerant) magnetism above $x > 0.05$ [41] and theoretical modeling that increasing frustration may suppress the bimagnon peak in the FeSCs [68].

To understand the nature of the magnetically ordered state as a function of doping more comprehensively, we turn to ARPES measurements below and above T_N , as shown in Fig. 3. By inspecting the two different high-symmetry cut band maps, presented in the form of second derivatives of energy distribution curves (EDCs), we can access the band

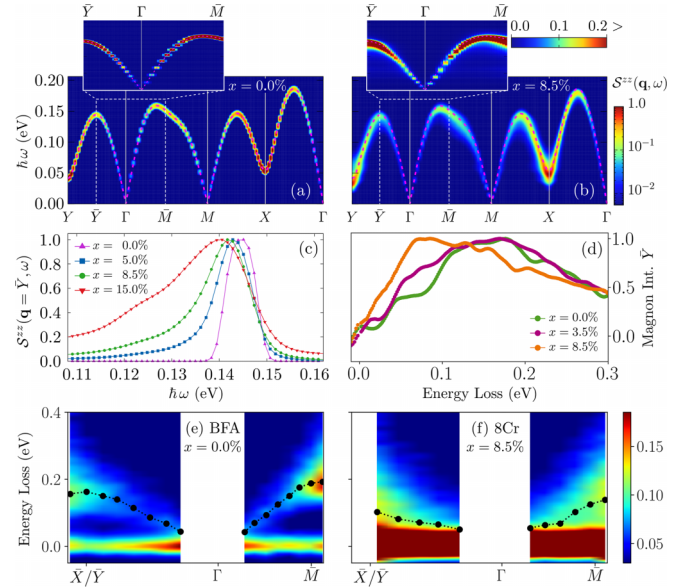


FIG. 4. (a), (b) Calculated dynamical structure factor of the J_1 – J_2 XXZ model for different doping levels (x). Insets show the RIXS probed region. The magenta dashed line marks the spin-wave dispersion for $x = 0$. (c) Structure factor at $\mathbf{q} = \bar{Y}$ for different x . (d) Experimental magnon at $\mathbf{q} \approx (0.5, 0) = \bar{X}/\bar{Y}$ for varying x after elastic and phonon contributions subtraction. RIXS momentum maps for (e) BFA and (f) Cr8.5%. Black dots are fitted ω_{peak} , as in Fig. 2(d).

maximum points and see the electronic bands' dispersion shape.

In the paramagnetic state, ARPES measurements of CrBFA reveal the increase in the hole pockets caused by Cr and the increasing electronic correlations, as characterized by the fractional scaling of the self-energy as a function of binding energy [24], a characteristic property of Hund's metals [2,30].

At $T = 20$ K, the BFA sample shows the typical $d_{xz/yz}$ orbital splitting from stripe-type itinerant order [Fig. 3(a)], reflecting the itinerant spin character in the AFM phase. The same reconstruction is present for Mn substitutions up to 8.5% along both ΓX and YZ cuts [17]. In contrast, for 8.5% Cr substitution [Fig. 3(b)], no such reconstruction is observed. Instead, the bands crossing the Fermi surface around X (mainly d_{xy}) become noticeably flatter, especially in the ordered state.

The absence of the electronic structure reconstruction in the case of CrBFA implies a more localized nature for the magnetic ground state at this doping, as also observed in previous experiments [41,70], and is consistent with the weaker orthorhombicity discussed above, since the d_{xz}/d_{yz} splitting is intrinsically tied to the structural and magnetic phase transitions. Motivated by these observations, we developed and solved a theoretical XXZ model of localized spins to calculate the effect of the doping disorder on the magnetic excitations' damping and dispersion, as detailed in the Supplemental Material [38].

Figure 4 shows theoretical results (dashed magenta line). Panels (a) and (b) present the dynamical structure factor, including intensities and linewidths (damping) as a function of Cr content x . We adopted $J_1 = 18.75$ meV to reproduce the magnon bandwidth at $x = 0$. The dashed white

rectangles mark experimentally accessible regions that are shown in more detail in the insets, wherein the color maps are rescaled to ease comparison with the experiment. Panel (c) shows the structure factor at $\mathbf{q} = \tilde{Y}$ versus energy for different x , while panel (d) presents, for comparison, the experimental magnon, after elastic and phonon subtraction, for the same $\mathbf{q}$ point. The finite width of the theoretical curve at $x = 0$ is due to the finite-size resolution of the calculations.

Our calculations explicitly include local fluctuations from Cr impurities and reproduce two key experimental observations: damping and softening of the excitations. The damping nearly doubles between $x = 0\%$ and 8.5% , matching the experimental trend. The suppression of the energy scale of the excitations (the softening), however, is stronger in experiments than in the theory, likely due to the missing itinerant-localized dual character of the spin excitations. This indicates that charge doping also contributes to magnon softening. Panels (e) and (f) show the experimental $\mathbf{q}$ dispersion maps for BFA and Cr 8.5% along the same high-symmetry direction as in insets of panels (a) and (b). Within the experimental $\mathbf{q}$ -range, theory and experiments agree with increasing spectral weight toward lower energies.

In addition to a good comparison with experiments, our calculations suggest that the magnon softening along the $(\pi, 0)$ direction is related to a shift of Fe-derived spectral weight toward the X/Y point with doping, as the magnetic excitations become more local, a $\mathbf{q}$ -region inaccessible to RIXS. This theoretical insight may also help explain a key feature of the CrBFA and MnBFA phase diagrams: the absence of HTSC. Although Cr concentrates spectral weight near $(\pi, 0)/(0, \pi)$, the associated excitations are highly incoherent (strongly damped), which is unfavorable for SC pairing [71,72].

Other substitutional studies also indicate that local fluctuations from Cr moments can dominate over charge doping effects, as observed in Cr substitution of Ni-doped BFA [73–75] and P-substituted BFA [76,77]. In CsFe $_2$ As $_2$, disorder alone may even enhance correlations [78,79].

Summary and conclusions. Our RIXS experiments investigated the evolution of the magnon dispersion along high-symmetry directions as a function of Cr content in CrBFA. Combined with theoretical modeling, our results suggest a scenario where, across the CrBFA x versus T phase diagram, disorder dominates the damping and partially explains the softening of the excitations. It reveals that charge doping mainly contributes to suppressing the excitations. This places disorder, instead of charge doping, as the primary effect in CrBFA, similarly to the case of Mn substitution, where ARPES observes minimal hole doping.

This is unexpected given the increasing electronic correlation, changing hybridizations, and electronic filling as observed by ARPES experiments of CrBFA [24]. Our results show that disorder overrides these electronic effects in governing spin dynamics. We thus expect that our work may stimulate theoretical/experimental work on the effects of disorder on Hund's metals.

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Data availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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