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Magnetic precursor temperature regimes in low-dimensional magnets provide a unique window into how spin, lattice, and electronic degrees of freedom become coupled before long-range spin order is established. Here we present a comprehensive investigation of the intertwined magnetic and lattice behavior above the Néel temperature (T_N) in the van der Waals (vdW) antiferromagnet CrSBr. The results reveal a crossover from high-temperature two-dimensional (2D) XY behavior dominated by short-range ferromagnetic (FM) fluctuations to an intermediate three-dimensional (3D) state dominated by interlayer antiferromagnetic (AFM) correlations preceding the onset of long-range AFM order. The heat capacity shows only the anomaly associated with AFM ordering, further indicating that this intermediate regime does not correspond to an additional thermodynamic phase transition. Polarized Raman measurements reveal phonon self-energy anomalies and relative intensity changes above T_N , indicating that the lattice dynamics have already renormalized in the magnetic precursor regime. These findings support the conclusion that dynamic spin-phonon coupling in CrSBr is already established prior to long-range AFM ordering and is mediated by short-range magnetic correlations.

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Since the emergence of two-dimensional (2D) magnets in 2017 [1,2], these materials have renewed interest in spintronics. Based on the Mermin-Wagner theorem [3–5], thermal fluctuations suppress long-range magnetic order in low-dimensional systems with continuous symmetries. However, magnetic anisotropy in 2D materials can stabilize magnetism,

allowing the realization of 2D magnets through mechanical exfoliation and motivating the exploration of novel van der Waals (vdW) layered crystals. More importantly, recently discovered 2D magnets are semiconductors offering a unique possibility for simultaneous control over charge and spin degrees of freedom, opening up exciting prospects for advancing novel spin-based technologies [6,7], such as polarized LEDs [8,9], nonvolatile memories [10,11], neuromorphic computing [12], etc. Among these, vdW layered CrSBr has emerged as a particularly promising candidate due to its strong coupling between magnetic, electronic, and lattice degrees of freedom [13,14], constituting a rich platform for exploring collective effects of charge, phonons, and magnetism.

The vdW CrSBr, characterized by a quasi-one-dimensional (quasi-1D) semiconducting electronic structure [15], is a layered A-type AFM semiconductor with a high Néel

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temperature of $T_N \approx 132$ K [16]. CrSBr exhibits a highly anisotropic lattice structure in which edge-sharing Cr-centered octahedra form chains with sulfur and bromine atoms occupying inequivalent positions above and below the Cr plane. Each rectangular layer is ferromagnetic (FM) with spins aligned parallel along the Cr chains, while adjacent layers are coupled antiferromagnetically through weak interlayer interactions. It exhibits triaxial magnetocrystalline anisotropy with a magnetic easy axis along the crystallographic b direction and experimental results from antiferromagnetic (AFM) resonance measurements reveal that the magnetic anisotropy in the ab plane even remains to some extent at temperatures above T_N [17]. Below T_N CrSBr exhibits temperature- and field-dependent spin-flip transitions from the AFM state to a fully aligned spin state [18].

CrSBr provides an appealing platform for studying low-dimensional magnetism, yet its critical behavior near the magnetic transition remains under debate. In particular, significantly different values have been reported for the critical exponent β , which describes how the magnetic order parameter (i.e., the sublattice magnetization) evolves when approaching T_N according to $M \propto (T_N - T)^\beta$. Second harmonic generation measurements on bilayer CrSBr yield $\beta = 0.36$, suggesting anisotropic Heisenberg behavior [19], while neutron powder diffraction gives a smaller value $\beta = 0.18$, lying between the expectations for 2D Ising ($\beta = 0.125$) and 2D XY ($\beta = 0.231$) models [20,21]. In contrast, single-crystal neutron diffraction reports $\beta = 0.231$, consistent with a 2D XY universality class. This discrepancy suggests that competing anisotropies and dimensional crossover effects play an important role near the magnetic transition.

The nature of the magnetic correlations above T_N is also under active discussion. Specific heat measurements have revealed, in addition to the sharp anomaly at T_N associated with the AFM transition, a broad feature around 155–160 K. This feature was initially attributed to a magnetic phase transition to a state in which spins align ferromagnetically within individual ab planes but randomly between adjacent ab planes [19,22]. This interpretation was later revised to be a crossover temperature scale below which the spins form short-range fluctuating patches both within and between ab planes [23].

Beyond its debated magnetic criticality, CrSBr has also emerged as a promising platform for investigating spin-lattice coupling. Recent Raman studies have shown that the magnetic ordering of CrSBr leaves clear fingerprints in the phonon response [24,25]. However, existing work has mainly emphasized signatures associated with the long-range ordered phase, while it remains unclear whether and how spin-phonon coupling occurs above the magnetic ordering temperature. This raises a broader question of whether the lattice can act as a sensitive probe of magnetic dynamics before long-range order is established. Demonstrating such dynamic spin-phonon coupling would reveal how fluctuating short-range spin correlations reshape phonon self-energies in a precursor regime, providing access to spin-lattice entanglement that is hidden from purely static probes.

Here we report a comprehensive study of the magnetic behavior and the accompanying strong dynamic spin-phonon coupling at temperatures above T_N of CrSBr single crystals. Temperature-dependent low-field susceptibility reveals a pro-

nounced 2D XY regime at elevated temperatures, driven by strong FM intralayer superexchange interactions. This view is consistent with small-angle neutron scattering results, which indicate that FM correlations persist up to temperatures well above 200 K [26]. With further cooling, this behavior evolves into a nondivergent response below $T \approx 160$ K, reflecting the increasing influence of weak AFM interlayer coupling. In addition, ac susceptibility measurements uncover the onset of anisotropic low-frequency dissipation below $T \approx 160$ K, with a dissipation maximum emerging near T_N and a rapid suppression of dissipation just below the transition. These features indicate that the characteristic temperature around 160 K does not correspond to a thermodynamic phase transition but rather signifies a crossover into a regime dominated by short-range intralayer and interlayer magnetic fluctuations. As summarized in Fig. 1(a), the magnetic behavior of CrSBr in the investigated temperature range follows a hierarchical evolution: from a high-temperature regime with 2D intralayer FM correlations to an intermediate 3D regime where interlayer AFM correlations become relevant and coexist with FM intralayer correlations, and finally to long-range AFM order below T_N . Importantly, the spin-phonon coupling is not limited to the long-range ordered phase below T_N , but is already evident in the intermediate temperature regime below ~ 160 K, as reflected by the pronounced anomalies in lattice dynamics that track the evolution of magnetic correlations. ac susceptibility measurements also reveal the existence of anisotropic low-frequency dissipation below T_N . This dissipation only exists when the field is applied along the b axis, which unequivocally identifies it as energy loss due to intralayer domain wall motion. Furthermore, the temperature dependence of the spin-flip field is exploited to probe the critical behavior of the staggered magnetization below T_N , providing additional information on the nature of the magnetic phase transition in CrSBr.

II. METHODS

The bulk high-quality CrSBr single crystals used in this study were supplied by HQ Graphene [27]. A Quantum Design magnetic property measurement system was used to investigate the magnetic properties of the CrSBr single crystals. Isothermal magnetization measurements were performed above and below the magnetic ordering temperature in magnetic fields up to 2 T. Temperature-dependent magnetization measurements were carried out over the temperature range 10–300 K in magnetic fields from 1 mT to 1 T. Temperature-dependent ac susceptibility measurements were performed using an ac magnetic field amplitude of 0.4 mT in the temperature range 50–160 K. Isothermal ac susceptibility measurements in superimposed dc magnetic fields were performed below the magnetic ordering temperature to investigate the temperature-dependent spin-flip magnetic field. The relatively high values of the susceptibility of CrSBr close to T_N require a correction for demagnetizing effects. The demagnetizing factors, estimated from the shape of the sample, are $N_a = 0.03$, $N_b = 0.17$, and $N_c = 0.8$ for the a , b , and c directions, respectively.

Temperature-dependent Raman spectra were collected using a Renishaw inVia confocal Raman microscope. A

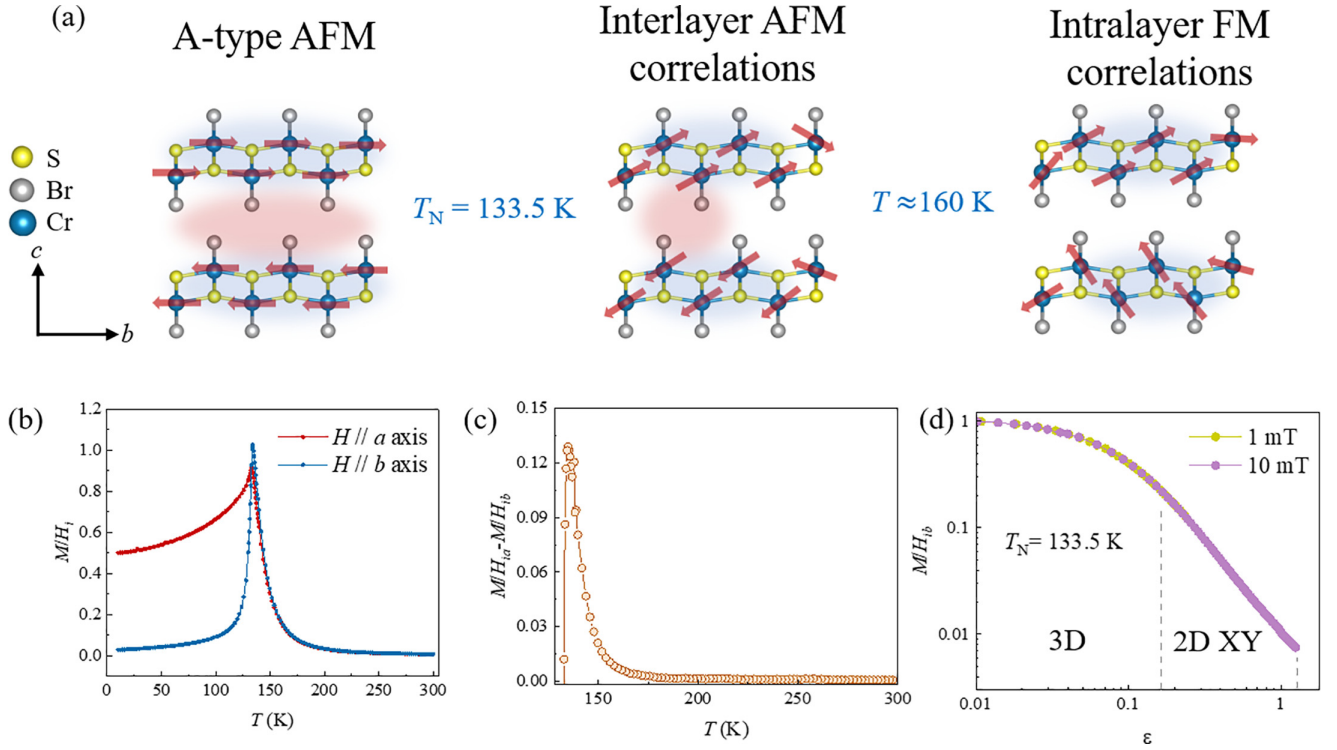


FIG. 1. Magnetic crossover and precursor dynamics in CrSBr. (a) Schematic illustration of the magnetic behavior across different characteristic temperatures (below 300 K) in CrSBr. (b) Susceptibility M/H_i versus temperature T applying the magnetic field H along the a and b axes. H_i is the internal magnetic field corrected for demagnetization effects ($H_i = H - NM$, with $N = N_a, N_b$, or N_c). (c) $M/H_{ib} - M/H_{ia}$ versus temperature. The applied magnetic field is $\mu_0 H = 10$ mT. (d) Linear susceptibility M/H_{ib} versus reduced temperature $\epsilon = (T - T_N)/T_N$ with $T_N = 133.5$ K at two applied magnetic fields 1 and 10 mT, respectively. The vertical dashed line marks $T \approx 160$ K, indicating the onset of short-range AFM correlations and 3D magnetic behavior.

frequency-doubled and linearly polarized Nd: YAG laser (532 nm; $E_L = 2.33$ eV) was used for excitation. Measurements were performed with a $100\times$ objective lens, corresponding to a laser spot size of approximately $1 \mu\text{m}$. The polarization of the reflected light was controlled using half-wave plates. The polarization direction of the incident light relative to the crystal axes was controlled by a motorized rotation stage, allowing the incident light to be aligned along both the a and b crystallographic directions, respectively. To ensure the reliability of the measured signals, the samples were freshly cleaved prior to each measurement. All measurements were repeated three times to minimize experimental uncertainty. The spectral data were fitted using a Voigt function. The peak position, intensity, and linewidth reported in this work correspond to the average values.

The temperature and magnetic field dependence of the specific heat was measured using a membrane-based nanocalorimeter operated in the ac steady-state mode, with a variable frequency to maintain a constant phase lag between the temperature oscillation and applied power [28]. In the central area of the calorimeter, the sample is placed on a 150-nm SiN_x membrane with a stack of thin film thermometers (NiCr/SiO_x), an ac heater (Cr), and an offset heater (Cr). The cryostat was kept below 100 mK while the sample temperature was controlled with the offset heater. A small amount of Apiezon-N grease was used to attach the crystal to the calorimeter. Correction for thermometer magnetoresistance

was applied. The specific heat of the background membrane and the grease were measured separately.

III. RESULTS AND DISCUSSION

We first focus on the low-field linear magnetic response of CrSBr. In the temperature range 10–300 K, this corresponds to an applied magnetic field of $\mu_0 H \leq 10$ mT. Figure 1(b) displays the in-plane magnetic susceptibility measured along the a and b axes. The transition to the AFM state is observed at $T_N = 133.5$ K. At first glance, the data suggest an isotropic in-plane response for $T > T_N$. However, as shown in Fig. 1(c), the magnetic anisotropy present below T_N persists above the transition up to $T \approx 160$ K. This observation is consistent with AFM resonance measurements, which demonstrate that anisotropy survives above T_N and does not disappear until temperatures around 180 K [17]. Having established that the in-plane susceptibility becomes effectively isotropic for $T \gtrsim 160$ K, we next examine whether its temperature dependence follows the expected behavior of a 2D XY model. Figure 1(d) shows the susceptibility as a function of the reduced temperature $\epsilon = (T - T_N)/T_N$ with $T_N = 133.5$ K, plotted on a log-log scale. In the temperature range 160–300 K, the susceptibility follows a power-law dependence $\chi \propto \epsilon^{-\gamma}$ with $\gamma \approx 1.71$, close to the value expected for a frustrated 2D XY system [29], and also near the 2D Ising exponent $\gamma = 1.75$. Below $T \approx 160$ K, the divergence is suppressed and the

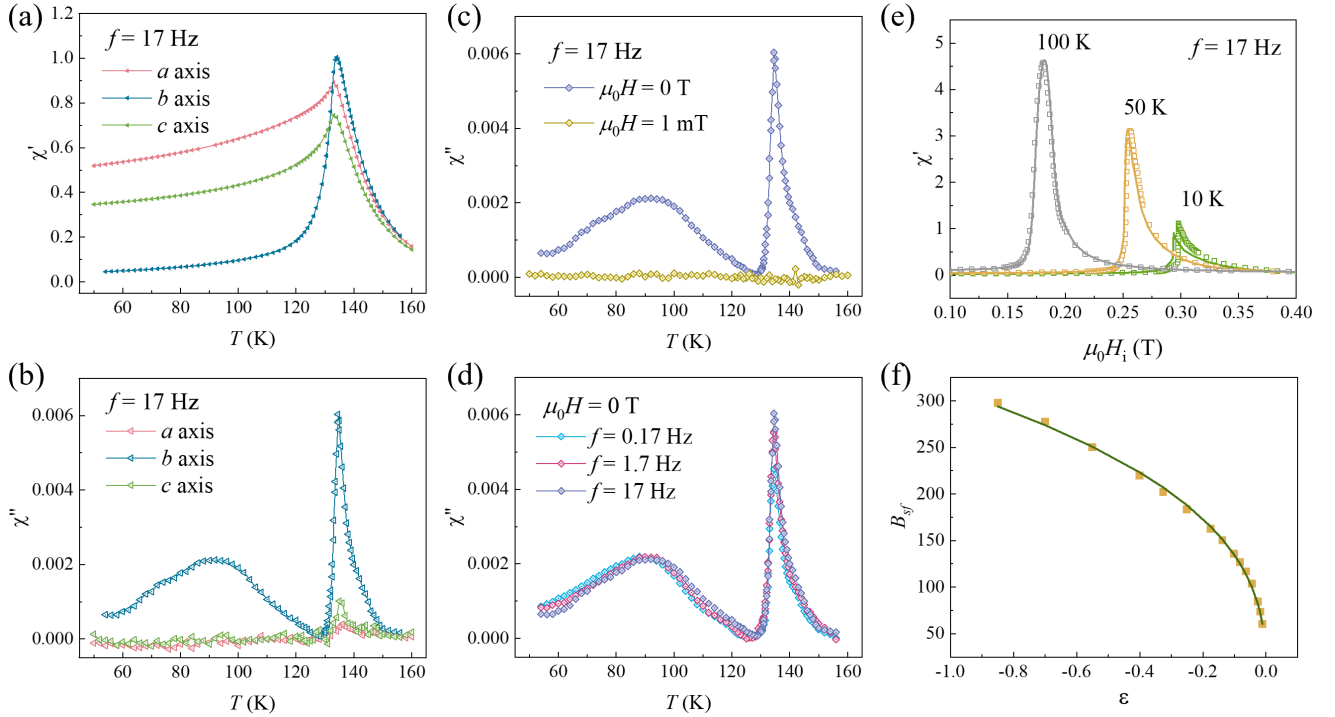


FIG. 2. Temperature and magnetic field-dependent ac magnetic susceptibility. (a) In-phase component χ' and (b) out-of-phase component χ'' of the ac susceptibility versus temperature T with $\mu_0 H_{AC} = 0.4$ mT applied along the a , b , and c axes. (c) Out-of-phase component of the ac susceptibility χ'' versus temperature T with $\mu_0 H_{AC} = 0.4$ mT applied along the b axis. The results are shown for $\mu_0 H = 0$ and 1 mT superimposed dc magnetic field. (d) Out-of-phase component of the ac susceptibility χ'' versus temperature T with $f = 0.17, 1.7$, and 17 Hz. (e) In-phase component of the ac susceptibility χ' versus internal dc magnetic field $\mu_0 H_i$ at 10, 50, and 100 K with the $\mu_0 H_{AC} = 0.4$ mT. (f) Spin-flip field B_{sf} versus reduced temperature ϵ with $T_N = 133.5$ K. Experimental data solid symbols and solid line fit to $B_{sf} \propto (-\epsilon)^\beta$. All ac susceptibility data have been corrected for demagnetization effects.

susceptibility deviates from the behavior of the power law. This crossover can be attributed to the onset of interlayer AFM coupling, which prevents the divergence of the low-field susceptibility. Importantly, the physical insight described here is different and more detailed compared to previous knowledge, where the crossover temperature scale separated a high-temperature PM spin state from a state where spins formed short-range fluctuating patches both within and between ab planes [23].

Several studies on CrSBr have reported an anomaly in the magnetic susceptibility below 40 K [20,30–32]. Based on muon spin relaxation measurements, the anomaly was assigned to slowing down of magnetic fluctuations below T_N , until a spin-freezing process occurs below 40 K [20]. A combined study involving magnetometry and magneto-transport measurements indicated that the Cr moments order ferromagnetically within each layer, but that defect moments remain unpolarized until becoming polarized below 40 K [32]. It was suggested that defect moments could arise from self-trapped electrons near donor sites (known as magnetic polarons [33]) rather than intrinsic magnetic impurities. However, later studies [18] using high-quality single crystals demonstrated the absence of this low-temperature phase, suggesting an extrinsic origin. This assumption was also corroborated by recent studies on annealed CrSBr crystals [34], demonstrating that the anomaly below 40 K is caused by FM CrBr₃ secondary-phase impurities. Notably, the anomaly in the magnetic susceptibility is not observed for

the CrSBr single crystals studied here, testifying to their high quality.

To further elucidate the nature of magnetic correlations and their dynamics, we turn to ac susceptibility measurements. Figures 2(a) and 2(b) capture the in-phase and out-of-phase components of the ac susceptibility measured at a frequency of $f = 17$ Hz with the ac magnetic field applied along the a , b and c axes. The temperature dependence of the in-phase component [Fig. 2(a)] again confirms that the magnetic anisotropy within the ab plane persists above T_N . The out-of-phase component reveals the onset of low-frequency relaxation below $T \approx 160$ K when the field is applied along the b axis, while no measurable dissipation is observed for fields along the a and c axes. The dissipation exhibits a pronounced peak near T_N , followed by an abrupt suppression to nearly zero upon entering the magnetically ordered state. This anisotropic dissipation indicates that it originates from short-range magnetic fluctuations with moments preferentially aligned along the $\pm b$ axis. A field applied along the b axis can couple with these fluctuations and induce energy dissipation, whereas fields along the a and c axes primarily generate reversible torque without significant energy loss. These fluctuations may remain confined within individual ab planes or become weakly correlated across layers through the interlayer AFM interaction. At temperatures below T_N , the out-of-phase component reemerges and develops a broad maximum below ~ 100 K, which is most naturally attributed to the motion of magnetic domain walls (see Fig. S1 in the Supplemental Material [35])

within the ab planes driven by the ac field applied along the b axis.

Figure 2(c) highlights the effect of superimposing a dc magnetic field along the b axis. The application of a dc field clearly eliminates the dissipation across the entire temperature range. Above T_N , short-range FM fluctuations, which in zero field are distributed between $\pm b$ orientations, are expected to align along the applied dc field. The disappearance of dissipation in this regime therefore suggests that the zero-field response originates from fluctuations involving oppositely oriented FM regions. This behavior is consistent with Kosterlitz-Thouless-type fluctuations, in which correlated regions with $\pm b$ -oriented moments form and dynamically interact. Correspondingly, the in-phase component of the ac susceptibility increases under a dc field, as confirmed experimentally [see Fig. S2(b) in the Supplemental Material [35]]. The disappearance of dissipation below T_N can be understood as follows: below T_N and in zero dc magnetic field, the system consists of magnetic domains with moments aligned along $\pm b$ within each ab plane, with AFM interlayer coupling enforcing opposite orientations in adjacent layers. The application of a dc field stabilizes single-domain states within each layer while preserving AFM stacking between layers. In this case, the in-phase susceptibility decreases under the applied dc field, in agreement with the experimental observations in Fig. S2(a) [35]. We emphasize that this discussion pertains to dc magnetic fields well below the spin-flip field. Consistent with this picture, the low field hysteresis measurements at $T < 100$ K show a finite opening at very low fields, which closes for fields above ~ 1 – 2 mT.

The out-of-phase component of the ac susceptibility measured at different excitation frequencies is shown in Fig. 2(d). The behavior above T_N approaching the AFM transition temperature is reminiscent of a critical slowing down of the spin dynamics. However, the experimental resolution does not allow a quantitative analysis of this behavior. Nevertheless, it is evident that the magnetic fluctuations responsible for the dissipation vanish abruptly upon entering the magnetically ordered state below T_N .

Below T_N , with the magnetic field applied along the b axis, CrSBr exhibits temperature- and field-dependent spin-flip transitions, as shown in Fig. S3 [35]. The temperature dependence of the spin-flip field was investigated using ac susceptibility measurements with superimposed dc magnetic fields. The amplitude of the ac field was 0.4 mT, such that $\chi(H)$ probes the differential susceptibility, i.e., the slope of $M(H)$. Figure 2(e) displays χ' as a function of $\mu_0 H$ for increasing and decreasing magnetic fields at three representative temperatures. The spin-flip field B_{sf} is defined from the maximum of $\chi'(H)$. The first-order character of the transition at low temperature is evident from the hysteresis between increasing and decreasing field sweeps. In addition, the spin-flip transition is accompanied by dissipation, as demonstrated by the field dependence of χ'' (Fig. S4) [35–37]. Single-spin scanning magnetometry measurements [38] have shown that the ferromagnetically aligned phase nucleates and coexists with the AFM phase during the transition. This phase coexistence indicates that the observed dissipation originates from irreversible domain wall motion. Figure 2(f) shows the temperature dependence of B_{sf} as a function of the reduced temperature

ϵ . Theoretical studies of easy-axis antiferromagnets, such as $(C_2H_5NH_3)_2CuCl_4$, have shown that the phase boundary between the AFM and field-aligned states, i.e., the spin-flip field, is proportional to the staggered (sublattice) magnetization [39]. Fitting the experimental data with $B_{sf} \propto (-\epsilon)^\beta$ using $T_N = 133.5$ K yields $\beta = 0.365$, in reasonable agreement with the expected value for a 3D XY universality class.

To better understand the phase transition behavior, we performed heat capacity (C_p) measurements under different magnetic fields as shown in Fig. S5 [35]. In zero field, a clear anomaly is observed at T_N manifested as a sharp peak in C_p , which is characteristic of a second-order phase transition associated with the AFM phase. With an increasing magnetic field, this transition anomaly is progressively suppressed. At 0.2 T, the peak becomes significantly weakened, indicating a spin-flip transition and that the external field destabilizes the long-range AFM order. This is also evident in Fig. S6 [35], showing the temperature dependence of the magnetic susceptibility for different magnetic fields applied along the b axis; the signature of the AFM phase transition is absent for applied magnetic fields $\mu_0 H \geq 0.2$ T. Turning back to the C_p results, a weak feature is observed around 155 K. However, considering its small magnitude and lack of systematic field dependence, we attribute this signal to a background contribution rather than to an intrinsic thermodynamic phase transition. In addition to this, no additional anomalies or latent-heat-like features were detected over the entire temperature range, suggesting the absence of other thermodynamic phase transitions. Therefore, the above-mentioned intralayer FM and interlayer AFM correlations should not be interpreted as distinct phase transitions. Instead, they are more appropriately understood as crossover phenomena associated with the gradual development of spin correlations or, equivalently, as correlation-driven magnetic precursors preceding the establishment of long-range magnetic order.

Polarized Raman spectroscopy serves as a powerful probe of anisotropic lattice vibrations, and its temperature dependence provides direct insight into the coupling between spin and phonon degrees of freedom. The Raman-active modes of CrSBr are known to belong to the A_g , B_{2g} , and B_{3g} symmetries [24]. However, in practice, only three out-of-plane layer-breathing A_g modes [see Fig. S7(a) [35]] are clearly observable due to the Raman selection rules imposed by the backscattering geometry. Figures 3(a) and 3(b) present the temperature-dependent Raman intensity maps of CrSBr measured with the incident polarization aligned along the a and b crystallographic axes, respectively. The waterfall-plot spectra of each axis are shown in Figs. S7(b) and S7(c) [35]. When the polarization is aligned along the a axis, the Raman response is dominated by A_g^2 while the other two modes are significantly suppressed. In contrast, in the b -axis configuration, three Raman-active modes are resolved at approximately 114 (A_g^1), 244 (A_g^2), and 342 (A_g^3) cm^{-1} at room temperature, originating mainly from vibrations of Br, Cr, and S atoms, respectively [40,41]. The distinct phonon contrast between the a and b axes highlights the pronounced in-plane vibrational anisotropy of the lattice.

Upon cooling, the spectral intensity of the A_g^1 and A_g^2 modes are progressively suppressed relative to that of the A_g^3 mode, as shown in Fig. 3(c) for the b -axis configuration, which likely

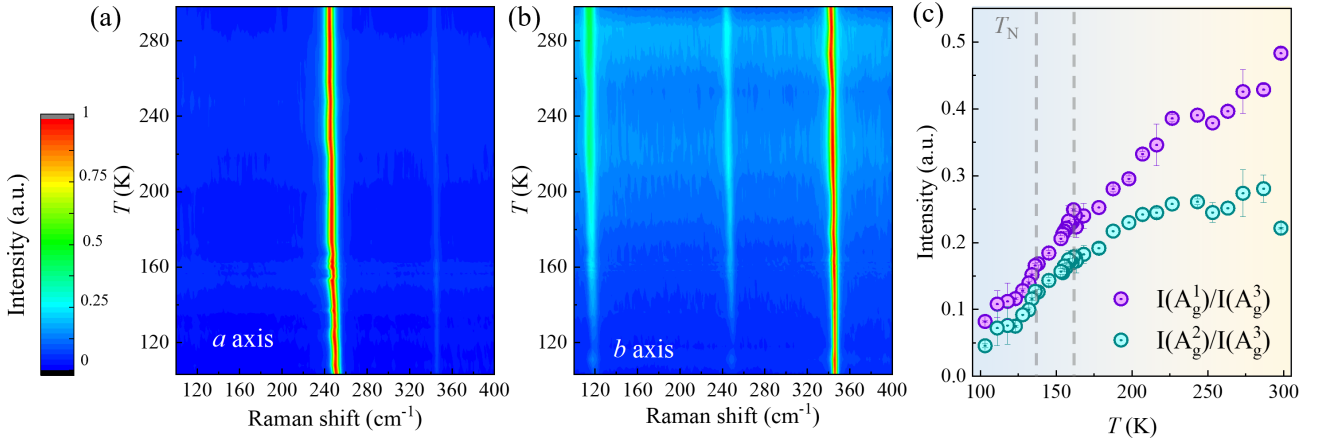


FIG. 3. Temperature-dependent polarized Raman response of CrSBr. (a) Raman intensity map measured in the *a*-axis polarization configuration. (b) Raman intensity map measured in the *b*-axis polarization configuration. (c) Temperature dependence of the intensity ratios of A_g mode along the *b* axis. The two gray dashed lines mark T_N and ~ 160 K, the latter indicating the onset of short-range AFM fluctuations.

indicates that the phonon response becomes increasingly anisotropic at low temperature. More specifically, in contrast to the nearly linear temperature evolution of the $I(A_g^1)/I(A_g^3)$ ratio, the $I(A_g^2)/I(A_g^3)$ ratio only exhibits an approximately linear behavior below 160 K and then gradually saturates upon further warming. This crossover is consistent with our previous observation that short-range interlayer magnetic fluctuations start to develop below 160 K. The Raman scattering intensity can be described by the formula $I \propto |\hat{e}_s^T R \hat{e}_i|^2$, where $\hat{e}_i$ and $\hat{e}_s$ are the polarization vectors of incident and scattered photons, respectively, and R represents the Raman tensor. The A_g symmetry here takes the general form

$$R(A_g) = \begin{pmatrix} R_{aa} & 0 & 0 \\ 0 & R_{bb} & 0 \\ 0 & 0 & R_{cc} \end{pmatrix} = \begin{pmatrix} ae^{i\phi_a} & 0 & 0 \\ 0 & be^{i\phi_b} & 0 \\ 0 & 0 & ce^{i\phi_c} \end{pmatrix}. \quad (1)$$

Here, a , b , and c denote the amplitudes of the Raman tensor elements along the principal crystallographic axes, while ϕ_a , ϕ_b , and ϕ_c represent their corresponding phase factors [42]. The Raman tensor thus reflects the crystal symmetry and is directly influenced by the electron-phonon coupling. Above 160 K, the short-range AFM interlayer fluctuations are suppressed, and the spin system evolves toward a 2D XY regime dominated by short-range FM fluctuations. Magnetic fluctuations induce a charge density difference and changes in the electric dipole moment or, equivalently, in the dielectric tensor components. Given the stronger Cr-vibrational character of A_g^2 , such spin fluctuations weaken the Raman tensor associated with this phonon mode. By contrast, the weaker sensitivity of the A_g^1 mode can be understood from its stronger interlayer character, which is mainly associated with the collective motion of the heavy Br atoms and is compared to the A_g^2 mode less coupled to the Cr spin network. Therefore, the distinct temperature dependence of these two intensity ratios can be understood as A_g^1 and A_g^2 exhibiting different levels of spin-phonon coupling that arises indirectly via electron-phonon coupling.

To quantitatively analyze the phonon behavior with higher precision, the individual Raman modes were fitted with Voigt functions to extract their linewidths and peak positions.

However, the temperature evolution of the full width at half-maximum [$\Gamma_{\text{exp}}(T)$] and the phonon frequency [$\omega_{\text{exp}}(T)$] in Fig. 4(a) is not purely monotonic and cannot be fully captured by conventional Klemens anharmonic decay [43], which accounts for the fact that a harmonic zone-center optical phonon acquires a finite lifetime due to anharmonic interactions and decays into two acoustic phonons via third-order anharmonicity. Using the standard anharmonic decay model, $\Gamma(T)$ and $\omega(T)$ were fitted using the following: $\Gamma_{\text{anh}}(T) = \Gamma_0 + A(1 + \frac{2}{\exp(\frac{\hbar\omega_0}{2k_B T}) - 1})$ and $\omega_{\text{anh}}(T) = \omega_0 + B(1 + \frac{2}{\exp(\frac{\hbar\omega_0}{2k_B T}) - 1})$. Here, Γ_0 and ω_0 denote the linewidth and phonon frequency in the zero-temperature limit, respectively, while A and B are adjustable parameters. $\hbar$ is the reduced Planck constant and k_B is the Boltzmann constant. For $\Gamma(T)$, in a purely anharmonic picture, a gradual decrease is expected upon cooling. Although the A_g^3 mode follows this behavior quite well throughout the temperature range, the A_g^1 and A_g^2 modes show slight deviations at low temperatures. However, these deviations are relatively weak and do not allow for a detailed analysis. Therefore, we primarily focus on the temperature dependence of the phonon frequencies.

The temperature dependence of the peak positions in Fig. 4(a) reveals a clear phonon hardening upon cooling for all three modes, as expected from the reduction of anharmonic phonon-phonon interactions at low temperature. Except for the A_g^3 mode, deviations from a smooth monotonic thermal trend already emerge around 160 K, well above T_N . This anomalous behavior is more pronounced for the A_g^2 mode than in A_g^1 , as evidenced by the residual phonon shift $\Delta\omega = \omega_{\text{exp}} - \omega_{\text{anh}}$ shown in Fig. 4(b). Other mechanisms such as electron-phonon coupling and lattice expansion that can induce a shift of phonon energies are less likely in this case given the large electronic gap and weak thermal expansion [20]. Therefore, the observed frequency anomaly is most naturally attributed to the coupling between lattice vibrations and short-range magnetic correlations. The frequency shift is proportional to the strength of the spin-phonon coupling (λ). This means that the renormalized phonon frequency can be described by

$$\omega \approx \omega_0^{\text{ph}} + \lambda \langle S_i \cdot S_j \rangle, \quad (2)$$

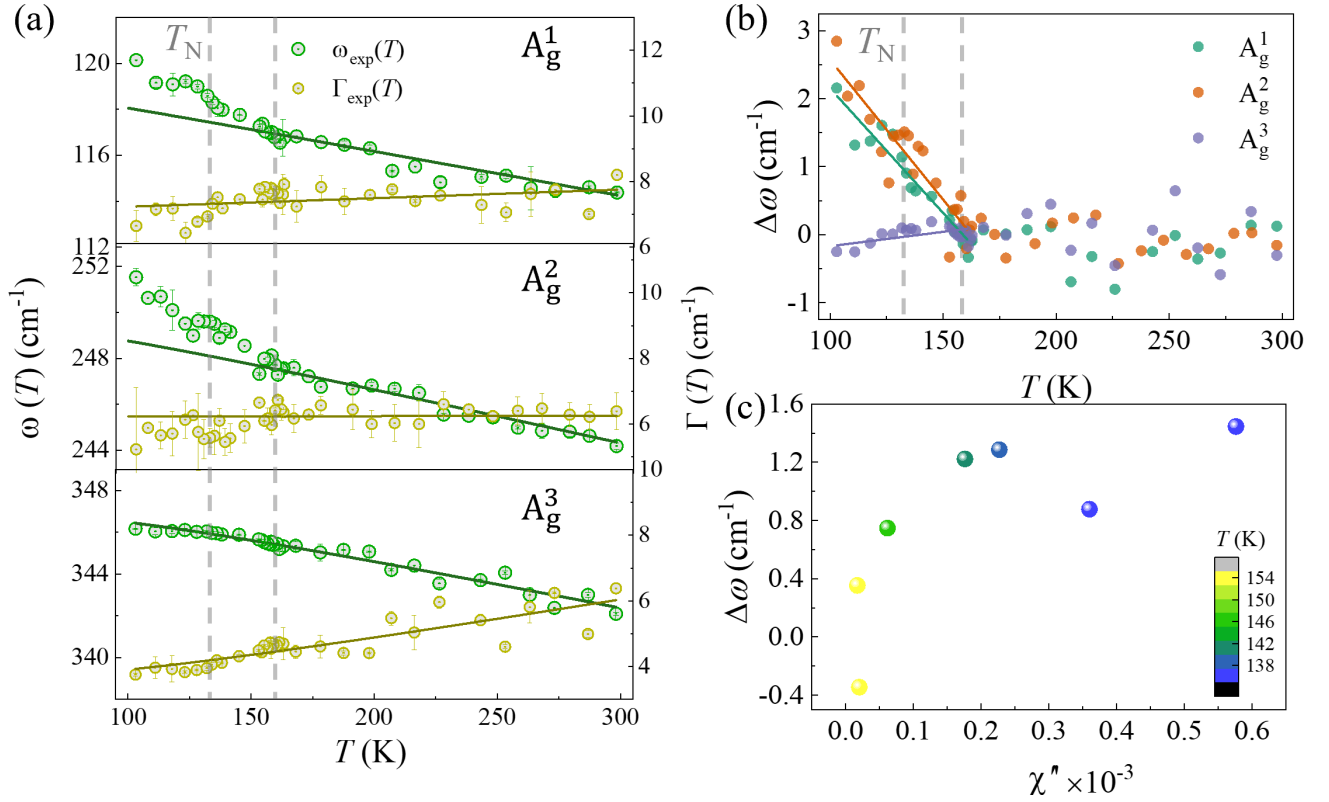


FIG. 4. Phonons renormalized by short-range magnetic fluctuations above T_N . (a) Temperature dependence of the fitted phonon frequency and linewidth for each A_g mode along the b axis. The temperature dependence of the phonon frequency (green solid line) and linewidth (yellow solid line) was fitted using the standard anharmonic decay model within the Klemens framework. (b) Residual phonon shifts $\Delta\omega = \omega_{\text{exp}} - \omega_{\text{anh}}$. The gray dashed lines mark T_N and ~ 160 K. (c) Correlation between the residual shift of the A_g^2 mode and the out-of-phase component of the ac susceptibility measured with the ac magnetic field applied along the b axis.

where S_i and S_j denote the nearest-neighbor spins located at the i th and j th Cr^{3+} sites [44,45]. Thus, the large frequency deviation of A_g^2 at low temperature indicates that A_g^2 is more strongly coupled to the magnetic degrees of freedom, which is consistent with the Raman intensity analysis discussed above. Importantly, the onset of the anomalous temperature dependence of the phonon frequency coincides with the temperature range in which short-range interlayer magnetic fluctuations become evident, as shown by the preceding analysis. It should be noted that these results complement previous Raman studies [42], which mainly emphasized Raman signatures associated with the long-range magnetically ordered phase. The result here indicates that the phonon self-energy is already renormalized by magnetic correlations prior to the establishment of long-range AFM order. A similar situation has been reported in the layered magnetic system $\text{Mn}_3\text{Si}_2\text{Te}_6$, where anomalous Raman self-energy effects persisting far above the magnetic ordering temperature were interpreted in terms of enhanced spin-phonon interaction associated with short-range magnetic correlations and competing short-range magnetic states [46]. In this scenario, the i th spin can be represented as

$$S_i = \frac{M}{4\mu_B} + \Delta S_i, \quad (3)$$

where M is the average sublattice magnetization per Cr^{3+} and ΔS_i is the spin fluctuation due to quantum and ther-

mal effects. The renormalization of the phonon frequencies above T_N can therefore be explained by short-range spin-spin correlations $\langle \Delta S_i \cdot \Delta S_j \rangle$ [47]. Figure 4(c) further shows the connection between the anomalous temperature dependence of the phonon frequency and the magnetic dissipation. The residual phonon shift of the A_g^2 mode exhibits a positive correlation with the out-of-phase component of the ac susceptibility. We may speculate that these short-range magnetic correlations most likely originate from interlayer AFM exchange interactions between neighboring Cr^{3+} spins. The absence of an A_g^1 anomaly in the intensity ratio but an anomaly here could arise because the A_g^1 frequency is more sensitive to spin-driven changes in the exchange pathways than to changes in the Raman-tensor response. Together, these results point to a strong dynamic spin-phonon coupling in CrSBr and further support the presence of short-range magnetic correlations above T_N .

Notably, the low-temperature deviations of the phonon frequency from the expectation of the standard anharmonic model exhibit opposite signs for different phonon modes. While the A_g^1 and A_g^2 modes lie above the extrapolation of the anharmonic model fit, the A_g^3 mode falls slightly below it, which is in agreement with previously reported results [24]. This indicates that spin correlations further harden the A_g^1 and A_g^2 modes, whereas they weakly soften the A_g^3 mode relative to the expectation of the standard anharmonic model.

The observed hardening and softening can be attributed to modulation of the magnetic exchange interaction by lattice vibrations, which can be associated with changes in bond lengths and angles along the relevant superexchange pathways [48]. A more detailed microscopic understanding of this effect requires further investigation.

IV. CONCLUSION

In conclusion, we identify the crossover near 160 K as from a magnetic perspective a gradual transition from a 2D XY behavior with short-range FM intralayer magnetic correlations to a 3D behavior including both FM intralayer and AFM interlayer magnetic correlations. ac susceptibility reveals that the precursor regime is dynamic and highly anisotropic, producing low-frequency magnetic dissipation only for fields applied along the b axis, with the dissipation strongly suppressed upon entering the ordered AFM state. The absence of a heat-capacity anomaly confirms that this feature is a magnetic crossover rather than a separate thermodynamic phase transition. Raman measurements further show that phonon self-energy anomalies develop in the same temperature range and correlate with the dissipative magnetic response. These results indicate that short-range magnetic dynamics renormalizes phonons above T_N , establishing dynamic spin-phonon coupling in the magnetic precursor regime of CrSBr. More broadly, our work advances the understanding of how cor-

related spin states above the magnetic ordering temperature can shape lattice dynamics in low-dimensional magnets, extending spin-phonon physics in CrSBr from the long-range ordered phase to the magnetic precursor regime.

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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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