

Algebraic Loop Liquid in the Pyrochlore CsNiCrF₆

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(Received 3 December 2024; revised 8 December 2025; accepted 22 January 2026; published 13 March 2026)

The β -pyrochlore CsNiCrF₆ is a charge ice—each tetrahedron of the pyrochlore lattice is occupied by two Ni²⁺ and two Cr³⁺ according to an ice rule, resulting in fully packed same-species loops with power-law length distribution. Using inelastic neutron scattering and numerical simulations, we show that the spin dynamics can be well described by a model with three antiferromagnetic interaction strengths, one for Ni–Ni, Cr–Ni, and Cr–Cr interactions. Because the strengths of the two intraloop interactions are quite different, the fluctuations of each type of loop can be identified in the spectrum. CsNiCrF₆ therefore forms an algebraic loop liquid in which power-law cation correlations control the spin dynamics. Our parameters predict a transition to spin nematic order with associated arrest of dynamics at $T_n \approx 0.55$ K, while we observe a freezing transition at $T_f = 2.3$ K. Measurements of magnetization, ac, and nonlinear susceptibility show that this is not a canonical spin glass transition so we associate it with the development of slow dynamics due to interloop nematic correlations.

DOI: 10.1103/xvxt-whns

Materials with two cations of different charge mixed on a geometrically frustrated sublattice experience charge frustration. The Anderson criterion [1,2] in which each plaquette has the same total charge is expected to identify ground state configurations for the cations. For the pyrochlore lattice with equal proportions of two cations, which we call “charge ice,” the criterion maps to ice rules for frustrated spins so the cation configurations have the same power-law (r^{-3}) correlation function characteristic of spin models supporting a Coulomb phase such as spin ice and

the pyrochlore Heisenberg antiferromagnet (PHAFM) [3,4]. Charge ice cation configurations also form fully packed loops in which each loop is composed entirely of one type of cation and every tetrahedron is visited by one loop of each type [5]. When both cations are magnetic, a simple model of the spin system has three interaction parameters, two for the like-ion interactions (intraloop) and one for the mixed-ion interaction (interloop); see Fig. 1(a) [6]. Evaluation of the energetics of a single tetrahedron suggests the model will have four zero-temperature ground states [see Fig. 1(b)], depending on the sizes of the magnetic moments and relative size and strength of the interactions [6]. The phases may be an ordered ferromagnet [I in Fig. 1(b)], partially ordered ferro- or antiferromagnets (II and III) reminiscent of “fragmented” phases [7,8], or a cooperative paramagnet with intraloop one-dimensional antiferromagnetic order and no interloop correlations (IV). Recent theoretical work that was confined to phase IV shows that the latter state undergoes a fluctuation driven order-by-disorder transition to a spin nematic [9]. At the transition, Heisenberg spins on the loops are replaced by emergent Ising degrees of freedom associated with the Néel

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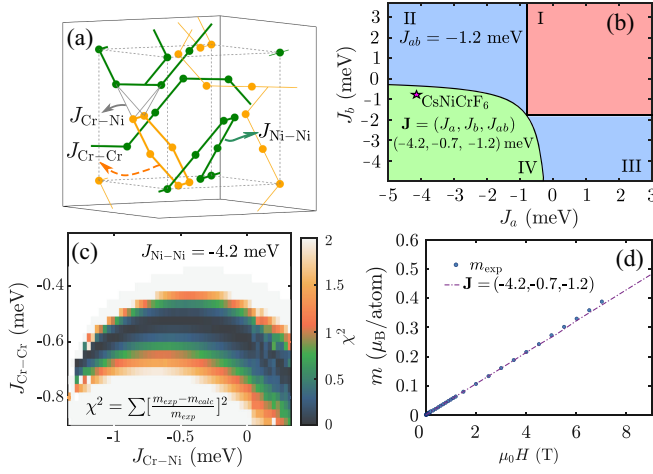


FIG. 1. Each tetrahedron of a magnetic charge ice has two ions of each type and we assign three interactions for the ion pairs in CsNiCrF_6 . Ions of the same type form intertwined closed loops that wander across the lattice and the tetrahedra are formed from a link of one loop of each type (a). This model has four classical ground states, of which IV (b) is an algebraic liquid of antiferromagnetic loops. In (b) we show the phase diagram in absolute units with spin lengths appropriate for CsNiCrF_6 (Refs. [6,9] have versions with relative units). DFT calculations suggested $J_{\text{Ni-Ni}} \sim -4$ meV, for which many combinations of $J_{\text{Cr-Cr}}$ and $J_{\text{Cr-Ni}}$ fit the magnetization well [e.g., (c) shows the region of well fitting parameters with $J_{\text{Ni-Ni}} = -4.2$ meV, which becomes more restricted as $J_{\text{Ni-Ni}}$ departs further from -4.2 meV]. The fit of the observed magnetization at $T = 0.7$ K for our optimal parameters, as finally determined by comparison with inelastic neutron scattering data, is shown in (d).

vectors of the loops. An important property of the nematic state is a distinctive slowing of the dynamics as these extended degrees of freedom order. The PHAFM is the point of the phase diagram where the four phases meet and approaching this point from within phase IV leads to stronger fluctuations and a higher nematic transition temperature.

Previous studies of CsNiCrF_6 have shown that it is a highly frustrated antiferromagnet with Curie-Weiss temperature $\theta_{\text{CW}} \sim -60$ K and no ordering down to low temperatures, but spin freezing at $T_f = 2.3$ K is attributed to a spin glass transition [10,11]. Later, a study of structural and spin correlations concluded that the cations form a charge ice, and that antiferromagnetic spin correlations are templated by this structure, making the system a multiple Coulomb phase [12]. The low-energy spin dynamics were found to follow various predictions for the PHAFM [13,14], suggesting universal aspects of dynamics in Coulomb phases. In this Letter we present inelastic neutron scattering measurements on CsNiCrF_6 that uncover higher-energy dynamics. We optimize the exchange parameters of a model charge ice to reproduce the results of magnetization, susceptibility, and inelastic neutron scattering experiments and thus interpret the spectrum as the signature of an algebraic loop liquid. Susceptibility measurements around

T_f show that the freezing is not a canonical spin glass transition. We propose that slow dynamics develop due to the partial realization of the nematic phase.

We have performed two inelastic neutron scattering experiments using the time-of-flight spectrometers LET [15] (ISIS, UK) and IN5 [16] (ILL, France), both equipped with “orange” helium cryostats. The sample was a single crystal of CsNiCrF_6 with mass ≈ 1.8 g, aligned with the (h, h, l) plane of reciprocal space horizontal. LET was operated with rep rate multiplication to provide incident energies of $E_i = 2.2, 4.1, 10.6$ meV simultaneously, with FWHM at the elastic line of 0.04, 0.11, 0.42 meV, respectively. Data were collected at 1.5 K by rotating the sample through 165° in steps of 1° for a total measurement time of approximately 24 h [17]. IN5 was operated in two settings: $E_i = 4.32, 20.45$ meV (FWHM at the elastic line of 0.12 and 1.35 meV, respectively) [18]. With $E_i = 4.32$ meV we rotated the sample through 150° in steps of 1° (10 min per point), with 0.5° steps in a 50° region centred on $(0, 0, l)$ (8.6 min per point), and because of strong spurious scattering we measured for equal times at 1.7 and 300 K to try to eliminate the background by fitting and subtraction of the spurious feature. With $E_i = 20.45$ meV we rotated the sample by 100° in 1° steps, counting for 14.4 min per step and measuring only at 1.7 K, the spurious scattering being folded into the elastic line in this setting. The data from both instruments were treated using the Mantid software to correct and normalize the raw data, and the Horace software to extract cuts from the four-dimensional $S(\mathbf{Q}, \omega)$. Using a small piece of CsNiCrF_6 crystal we measured the susceptibility and magnetization in field up to 7 T and down to 1.5 K using a SQUID magnetometer (MPMS-XL) at PSI. We investigated ac susceptibility and field-cooled and zero-field cooled magnetization using the miniature dilution refrigerator and SQUID magnetometer developed at the Institut Néel [19]. Extensive investigations of the nonlinear susceptibility, accessed by measuring the magnetization in small fields, were made using both instruments.

In Figs. 1(c) and 1(d) we show our initial parametrization of the spin Hamiltonian of CsNiCrF_6 . Density functional theory (DFT) calculations provide an initial estimate of the parameters [20], with $\mathbf{J} \equiv (J_{\text{Ni-Ni}}, J_{\text{Cr-Cr}}, J_{\text{Cr-Ni}}) = (-4.5, -0.6, 0.5)$ meV. $J_{\text{Cr-Ni}}$ has a large spread of values that may be either ferromagnetic or antiferromagnetic, while $J_{\text{Ni-Ni}}$ and $J_{\text{Cr-Cr}}$ are firmly predicted to be antiferromagnetic. This can be attributed to the greater variation of distances and angles among mixed-cation pairs and F^- anions upon relaxation, as compared to same-cation pairs. The interactions are strong enough to preclude approaching the saturation magnetization with typical laboratory fields, and when we survey parameters in the region of the estimates from DFT by fitting the magnetization using a single-tetrahedron calculation with $S_{\text{Ni}} = 1$,

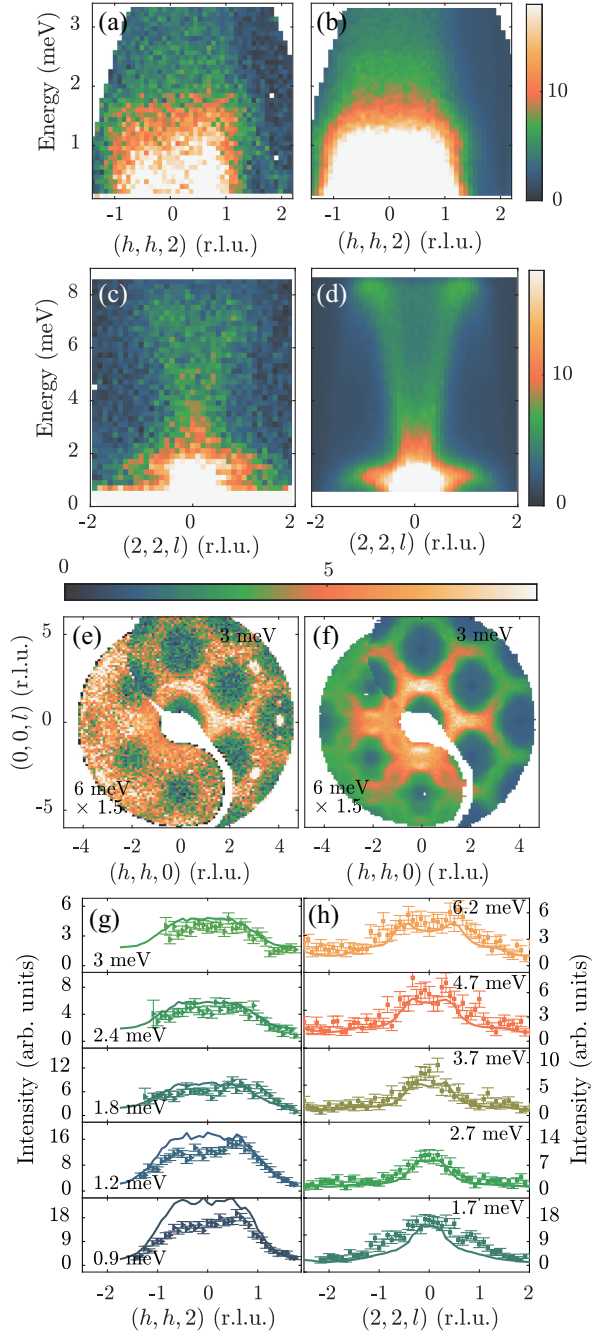


FIG. 2. Measurement and simulation of the inelastic neutron spectrum of CsNiCrF_6 . There is significant spectral weight below 2 meV (a), and also weaker scattering extending up to about 10 meV (c). This higher-energy scattering has a distinctive structure factor with pinch points that appear to broaden at higher energies as the intensity disperses outward from the pinch points [(c) and (e)]. Simulations of the scattering using numerical spin dynamics reproduces these features [(b), (d), and (f)] and scales well onto cuts through the structure factor [in (g) and (h) points show experimental data measured with two incident energies, lines show the simulated structure factor integrated in the same ranges as used for the experimental data]. All experimental data are from LET.

$S_{\text{Cr}} = 3/2$, we find a domain of parameters in which both antiferromagnetic and ferromagnetic $J_{\text{Cr-Ni}}$ can fit the magnetization well [Figs. 1(c) and 1(d)] [20]. The inverse susceptibility turns downward below the Curie-Weiss regime, and, from Monte Carlo simulations of magnetic charge ice, it is clear that this behavior is associated with antiferromagnetic $J_{\text{Cr-Ni}}$ [20]. Our parameters become $\mathbf{J} = (-4.5, -0.65, -1.15)$ meV.

We next explore the form of $S(\mathbf{Q}, \omega)$, in the (h, h, l) plane of reciprocal space. In cuts along nodal lines a broad feature disperses weakly up to ≈ 2 meV [Figs. 2(a) and 2(c)], and elsewhere in reciprocal space the signal has a quasielastic Lorentzian form in the energy transfer. In these respects we recover previous results, which also showed that at $\hbar\omega = 0.5$ meV the structure factor resembles that of the PHAFM with broadened pinch points [12], we refer to this energy range as “low” energy. However, the data presented here show there is also intensity above 2 meV. This “high” energy scattering is weaker than the low energy features just mentioned, but it can be clearly seen in slices along high symmetry directions, where a broad column of intensity rises from pinch point positions [Figs. 2(a) and 2(c)], and in constant energy cuts, where the PHAFM structure factor is increasingly broadened at higher energies [Fig. 2(e)]. The intensity disperses outward from the pinch points, tending to fill in the holes in the structure factor [Figs. 2(e), 2(g), and 2(h)] toward the top of the feature, which is at ~ 8 meV.

Using Langevin dynamics in the package Sunny [38] we can simulate $S(\mathbf{Q}, \omega)$ to compare with the experimental data. We used a system of 12^3 pyrochlore cells in which the pyrochlore lattice was populated with a charge ice configuration of Ni^{2+} and Cr^{3+} drawn in an unbiased way from all ice rule obeying configurations, giving an r^{-3} dependence to the cation correlations from which power-law loop length statistics result [5,6,9]. No ice-rule defects or other disorders were considered [9,20]. We refer to such systems as “ideal” charge ices. Using the preliminary parameters for the different interactions produced the distinctive low and high energy dispersive features, whose bandwidths are primarily controlled by $J_{\text{Cr-Cr}}$ and $J_{\text{Ni-Ni}}$, respectively. $J_{\text{Cr-Ni}}$ was found to redistribute intensity, particularly around the pinch points, whose relative sharpness at low energies seems to be maintained by $J_{\text{Cr-Ni}}$ before they are broadened away at higher energy [20]. Optimizing the parameters to match the observed bandwidths and spectral weight distribution confirms that $J_{\text{Cr-Ni}}$ is antiferromagnetic and results in $\mathbf{J}_{\text{opt}} = (-4.2, -0.7, -1.2) \pm 0.1$ meV, which can be seen to lie in the cooperative paramagnetic phase IV in Fig. 1(b). Cuts in the simulated data are compared with their experimental counterparts in Figs. 2(b), 2(d), and 2(f)–2(h), and previous low energy data from [12] in [20]. The model reproduces the spectrum, magnetization, and Curie-Weiss temperature [20] well,

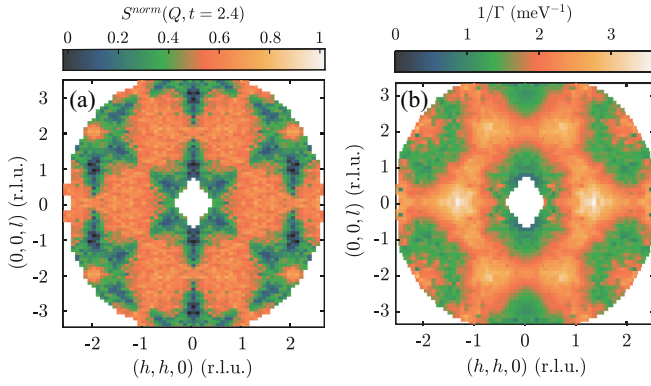


FIG. 3. Momentum dependence of spin relaxation rate obtained from simulations as the normalized relaxation time $S^{\text{norm}}(Q, t = 2.4) = S(Q, t = 2.4)/S(Q, t = 0)$ [13,14,20] (a) and from experimental data as the inverse width of Lorentzian fits to the quasielastic response [(b), data measured at IN5]. The experiment and simulation are performed at 1.6 K; the exchange parameters for the simulation are those obtained in the main text.

while other suggestions for the cation structure (i.e., random, charge ice with defects, or charge ordered) do not [20], implying that the magnetic interactions, correlations, and dynamics are indeed controlled by an underlying loop structure of the cations with power-law length distribution. We therefore call this state an “algebraic loop liquid” and propose that it will be the basis for understanding the magnetism of β -pyrochlores where charge ice structures are anticipated.

As in the PHAFM, we may try to classify the fluctuations by wave vector and discern something about the emergent dynamics of the algebraic loop liquid [13,14]. In simulations of ideal algebraic loop liquids (see Ref. [20] for details) a qualitatively similar wave vector dependence of the spin relaxation rate to the PHAFM is obtained: fast relaxation along nodal lines; generic regions of slower, wave vector independent relaxation delineated by the nodal lines; and the slowest relaxation at the intersection of the nodal lines; see Fig. 3(a). The fast relaxation process in the PHAFM is due to spin wave excitations living on the correlated spin structure [13]. In the algebraic loop liquid the scaling of the density of states, which has a one-dimensional character, suggests that such excitations are also confined to the loops [9]. This is clearly seen for CsNiCrF₆, where the dispersion maxima of the two branches agree well with those expected for antiferromagnetic chains with $J_{\text{Ni-Ni}} = 4.2$ meV and $S = 1$ or $J_{\text{Cr-Cr}} = 0.7$ meV and $S = 3/2$, at 8.4 and 2.1 meV, respectively, as can be seen in Figs. 2(c) and 2(d).

The two slower relaxation processes in the PHAFM originate from the solenoidal character of the spin correlations in the magnetic Coulomb phase, and the wave vector and temperature dependence of the relaxation rate reflects fluctuations in which the nondivergence criterion of the magnetic Coulomb phase is either conserved or violated (spin diffusion and relaxational dynamics at small and

generic wave vectors, respectively) [13]. The qualitative similarity of the wave vector dependence of the simulated relaxation rate shows that ideal loop liquids share these dynamics. The root of this similarity is that the PHAFM explores a continuously connected manifold of states [39,40] in which all possible looplike spin correlations and their intra- and interloop fluctuations may be instantaneously identified. The algebraic loop liquid inherits the same power-law spin correlation function from the charge ice and explores only the subset of states containing the physical loops imposed by the template [9]. Nonetheless, cooperative fluctuations mediated by the interloop interaction, acting alone or in tandem with the intraloop interactions, may occur, giving the same types of fluctuation with respect to the emergent gauge structure. This is why we see the role of $J_{\text{Cr-Ni}}$ in distributing nondispersive intensity in regions not dominated by intraloop fluctuations, and why, despite the strong bond disorder, the structure factor of CsNiCrF₆ at low energy closely resembles that of the PHAFM; see Ref. [20] for further discussion.

However, when we extract the relaxation rate for CsNiCrF₆ using the width of the quasielastic response in our experimental data [20], we find that the strictly wave-vector-independent relaxation rate in the generic regions is less marked, and the nodal lines and slow-relaxation regions, though present, are much less sharply defined; see Fig. 3(b). The wave vector dependence of the relaxation rate in CsNiCrF₆ implies that while real dynamics are closely related to those of an ideal loop liquid, the mismatch with the simulation is a first indicator that we have not completely described the physics of CsNiCrF₆, particularly at very low energy, where indeed the largest mismatch between experiment and simulation can be seen in Fig. 2. A second one is the spin freezing transition, which is also associated with the longest experimental timescales or lowest energy fluctuations. In simulations of ideal systems, the loop liquid develops on cooling, followed by spin nematic ordering at T_n . Spin freezing, as measured by the Edwards-Anderson parameter, does not occur in simulated ideal loop liquids until slow dynamics develop at the nematic transition. Using $\mathbf{J}_{\text{opt}}$ we predict that $T_n = 0.55$ K in CsNiCrF₆, but the experimental heat capacity shows no sign of a phase transition down to 0.3 K, only a broad peak centered at T_f [20].

At T_f the magnetization becomes history-dependent [Fig. 4(a)], and frequency and temperature dependent peaks appear in χ' and χ'' [e.g., Fig. 4(c)] as at a canonical spin glass transition [41,42]. The Mydosh parameter φ [43,44] extracted from the frequency and temperature dependence of the peak in χ' [Fig. 4(d)] is 0.014 ± 0.001 , close to the range known for canonical spin glasses ($0.004 < \varphi < 0.018$) and well below the range known for cluster glasses ($0.03 < \varphi < 0.06$). This value of φ implies a cooperative freezing, but is not uniquely diagnostic of a spin glass. On the other hand, T_f increases with applied field [Fig. 4(b)], while in a canonical spin glass T_f follows a

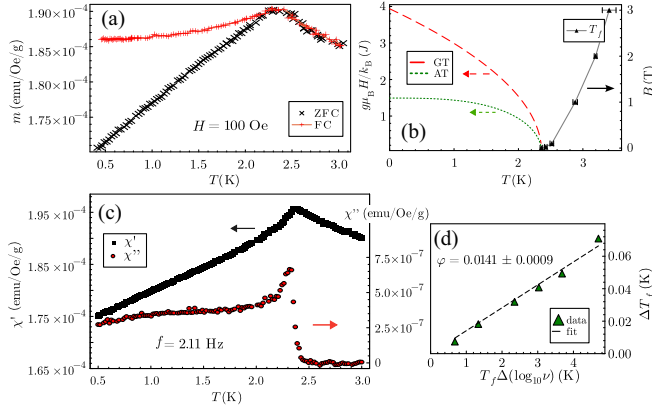


FIG. 4. (a) Susceptibility characterization of the freezing transition: example of the field-cooled (FC) and zero-field cooled (ZFC) magnetizations. (b) Field dependence of T_f , as given by the temperature of the cusp in the ZFC magnetizations, compared with the Gabay-Toulouse (GT) and de Almeida-Thouless (AT) lines. (c) Example of the ac susceptibility. (d) Extraction of the Mydosh parameter (ϕ) from the peak position of χ' where the shifts of the peak $\Delta(\log_{10} \nu)$ and ΔT_f are with respect to the lowest frequency measurement (0.21 Hz), which is in close agreement with the smallest field ZFC magnetization measurement.

decreasing trend with increasing field such as the Gabay-Toulouse or de Almeida-Thouless lines [also shown in Fig. 4(b)] [45–48]. Lastly, in CsNiCrF_6 the nonlinear susceptibility—a vital characteristic of a spin glass transition [49,50]—cannot be detected close to T_f [20]. These facts speak against the interpretation of CsNiCrF_6 as a spin glass.

We speculate that the noncanonical freezing observed in CsNiCrF_6 is another signature of the restricted manifold of states, which, being no longer continuously connected, gains a more complex landscape than the PHAFM [51]. Nematic correlations [52] and associated slow dynamics develop as the antiferromagnetic correlation length in the loops exceeds a threshold value [9], but the system is then arrested in the corrugated energy landscape, and the true nematic transition is not realized [51,53–55]. We note that freezing anomalies of diverse forms are observed at $20 < \theta_{\text{CW}}/T_f < 55$ in β -pyrochlores in which both cations are magnetic [56–60] (e.g., classical spin glass in CsNiFeF_6 , weak freezing in CsMnFeF_6), while in those with only one magnetic cation (e.g., CsZnFeF_6) freezing has not been observed at the temperatures so far investigated, and the much larger value of $\theta_{\text{CW}}/T_f \sim 120$ is only a lower bound. This situation is rationalized by the charge ice picture—strong intraloop interactions can produce the reasonably large Curie-Weiss temperatures observed in all the compounds, but interloop interactions are required to produce the magnetic anomalies universally denoted as spin freezing. However, the freezing transition temperature may be enhanced by effects not included in our model (e.g., distributions of exchange strengths as hinted at by our DFT calculations of $J_{\text{Cr-Ni}}$, or local spin anisotropies), and further studies are required to identify how such effects

control the low energy fluctuations and hence both the nature of the freezing transitions and wave vector dependence of the relaxation rate.

In conclusion, we have surveyed $S(\mathbf{Q}, \omega)$ in CsNiCrF_6 and two contributions to the spin dynamics with dispersive components both at low energy ($\hbar\omega < 2$ meV), and extending up to higher energies ($\hbar\omega < 10$ meV). Using numerical spin dynamics simulations and fitting to bulk quantities we find that a model with $\mathbf{J} = (-4.2, -0.7, -1.2) \pm 0.1$ meV reproduces the scattering, magnetization, and susceptibility signals. This model is based on unbiased application of the ice rule to generate the underlying bond disorder, which results in fully packed loops of either nickel or chromium, on which strong spin correlations develop, so CsNiCrF_6 is an algebraic loop liquid. This state of matter encourages further theoretical and experimental study—there are many β -pyrochlores [56–58], and this Letter shows that the charge ice model may rationalize their magnetic properties. It would be worthwhile to reinvestigate the classification of many of them as spin glasses and seek direct experimental evidence of nematic correlations. Further, one may ask if all the phases in the diagram of Fig. 1(b) can be realized, and if other novel cooperative behavior beyond that envisaged in Ref. [6] can be discovered. One interesting possibility is a compound with parameters closer to the PHAFM point of the phase diagram, where nematic fluctuations are stronger and might promote the nematic ordering transition above the freezing temperature; another would be examples where quantum effects are strong and the system becomes fully packed loops of quantum spin chains [61].

Note added—We recently learned of a preprint by Pohle and Shannon [62], in which biquadratic interactions are proposed to produce spin nematic phases and unconventional freezing in pyrochlore antiferromagnets such as $\text{NaCaNi}_2\text{F}_7$ [63]. Biquadratic interactions are of particular importance in the case of $S = 1$ and so could be another relevant extension of our model of CsNiCrF_6 .

Acknowledgments—We are pleased to acknowledge discussions with Martin Mourigal, Markus Müller, Collin Broholm, and Ch. Rüegg, and the latter for supervision of A. H. Neutron scattering experiments were carried out at ISIS [17], UK; the high flux reactor of the Institut Laue Langevin in Grenoble [18], France; and the continuous spallation neutron source SINQ at the Paul Scherrer Institut at Villigen PSI in Switzerland. Work at PSI was partly funded by the Swiss National Science Foundation grant “New structures and dynamics emerging from frustration” (200020_182536). The research leading to these results has received funding from the European Union’s Horizon 2020 Research and Innovation Programme, under Grant Agreement No. 824109 (MicroKelvin). N. C. acknowledges partial support from the NCCR MARVEL, a National Centre of Competence in Research, funded by the Swiss National Science Foundation (Grant No. 205602).

Data availability—The data that support the findings of this article are openly available [17,18,64]; embargo periods may apply.

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