

Dielectric signatures of crystal-field and low-temperature correlated dynamics in NdMgAl₁₁O₁₉

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We report dielectric spectroscopy of single-crystalline NdMgAl₁₁O₁₉, a magnetoplumbite hexaaluminate in which localized Nd³⁺ moments coexist with a polarizable AlO₅ bipyramidal network. The real part of the permittivity $\epsilon'_c(T)$, measured along the crystallographic c axis, increases as the temperature is lowered from 275 K to 30 K and is frequency-independent between 4 Hz and 50 kHz. At lower temperatures, a frequency-dependent decrease in permittivity is observed, followed by a further upturn below 2 K. The high-frequency $\epsilon'_c(T)$ is described by a Barrett formula supplemented by an effective two-level contribution, yielding a robust gap of $\Delta = 25.9(3)$ K consistent with the lowest Nd³⁺ crystal-electric-field (CEF) splitting. Below ~ 30 K, the dielectric response becomes strongly frequency and magnetic-field dependent. Isothermal $\epsilon'_c(H)$ measurements reveal a reproducible low-field crossover near $\mu_0 H_c \simeq 0.85$ T, which we attribute to the competition between antiferromagnetic correlations and Zeeman splitting of the ground-state Kramers doublet. NdMgAl₁₁O₁₉ thus provides a Kramers reference system in which dielectric signatures of the excited-state CEF manifold can be distinguished from those of the field-tuned, correlation-dominated ground-state doublet in the centrosymmetric frustrated magnetoplumbite host.

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Introduction. Rare-earth frustrated magnets provide a natural setting in which geometric frustration, strong spin-orbit coupling, and crystal-electric-field (CEF) effects combine to produce highly anisotropic low-energy degrees of freedom on frustrated lattices [1–5]. In such systems, bulk response functions may depend not only on exchange interactions but also on the structure of the CEF manifold and its coupling to lattice and orbital degrees of freedom. This broader view is supported by studies of rare-earth oxides and magnets in which CEF excitations interact with phonons and other low-energy modes, producing observable anomalies in optical and magnetodielectric response [6–8]. Dielectric measurements can therefore provide information complementary to conventional magnetic probes when local electronic and lattice degrees of freedom are significantly coupled. The magnetoplumbite-derived hexaaluminate family REMgAl₁₁O₁₉ is particularly well suited for examining this interplay because frustrated rare-earth magnetism coexists with a polarizable oxide framework [9–11]. Although the average structure is centrosymmetric, local off-centering within oxygen bipyramids and the resulting dipolar frustration generate a soft dielectric

background [11–16]. The nonmagnetic member EuAl₁₂O₁₉ establishes this dipolar host background in the absence of rare-earth CEF physics [11]. Recent studies on magnetic members of the family show that the background is not independent of the rare-earth ion: in CeMgAl₁₁O₁₉, a broad field-dependent dielectric anomaly develops on top of the Barrett-like background and reflects the low-energy Kramers manifold [10], whereas in PrMgAl₁₁O₁₉, local symmetry lowering and non-Kramers physics produce a qualitatively different low-temperature dielectric response [9]. Taken together, these results indicate that the dielectric response of this family provides a sensitive probe of how the single-ion CEF scheme is expressed through the polar host lattice.

NdMgAl₁₁O₁₉ provides a particularly informative Kramers case within this series. Nd³⁺ is a Kramers ion with $J = 9/2$, and previous magnetic and thermodynamic work identified NdMgAl₁₁O₁₉ as a quasi-two-dimensional triangular-lattice magnet with a strongly anisotropic ground-state doublet, weak antiferromagnetic interactions ($\Theta_{CW} \approx -0.5$ K), and no conventional long-range order down to the millikelvin regime [17]. The same work, supported by point-charge analysis of the Nd³⁺ crystal-field scheme [18], established a low-energy CEF hierarchy distinct from those of its Ce and Pr analogues, placed the first excited doublet near $E_1 \simeq 2.17$ meV (≈ 25.2 K), and identified from Schottky analysis of the field-evolving low-temperature thermodynamics a characteristic low-field scale of approximately 0.75–1 T associated with the ground-state doublet. A central question is therefore how these Nd-specific energy scales appear in the dielectric response.

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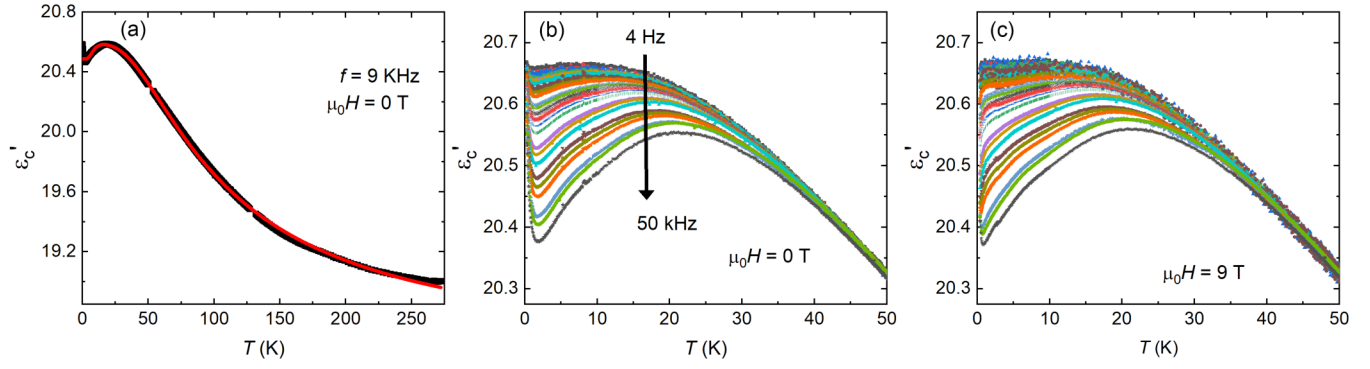


FIG. 1. Dielectric response of NdMgAl₁₁O₁₉. (a) Zero-field permittivity $\varepsilon'_c(T)$ measured at $f = 9$ kHz; the solid line is the combined fit using a Barrett background and a term associated with thermal depopulation of the first excited CEF doublet [Eqs. (1)–(3)]. (b) and (c) Temperature dependence of $\varepsilon'_c(T)$ at selected frequencies in (b) $\mu_0 H = 0$ and (c) $\mu_0 H = 9$ T.

Here we report dielectric spectroscopy of single-crystalline NdMgAl₁₁O₁₉ over broad temperature, frequency, and magnetic-field ranges. We identify three superposed contributions to the permittivity: a Barrett-like host background, a CEF-related contribution from the first excited doublet, and a low-temperature dispersive channel governed by the interplay of antiferromagnetic correlations and Zeeman splitting of the ground-state doublet.

Results. Single crystals of NdMgAl₁₁O₁₉ were grown by solid-state reaction followed by optical floating-zone growth, as described in Refs. [17,19–21]. Dielectric permittivity was measured along the c axis using an Alpha-AN impedance analyzer (Novocontrol Technologies) in a ³He cryostat (0.3–275 K, $\mathbf{H} \parallel c$, up to 9 T); further details of the measurement protocol follow Refs. [9,10].

Figure 1(a) shows the temperature dependence of the real part of the dielectric permittivity, $\varepsilon'_c(T)$, measured in zero magnetic field at $f = 9$ kHz up to 275 K. On cooling, $\varepsilon'_c(T)$ increases, passes through a broad maximum near ~ 25 –30 K, then decreases and finally shows a further upturn below a few kelvin. Between approximately 35 and 200 K, the response is nearly frequency-independent within the experimental resolution, indicating that this interval is dominated by the intrinsic high-frequency dielectric background. In this temperature range, the smooth increase of $\varepsilon'_c(T)$ on cooling is well described by a Barrett formula [Eq. (1)], as commonly used in incipient ferroelectrics and quantum paraelectrics, and may reflect weak softening of a polar phonon.

$$\varepsilon'_{\text{Barrett}}(T) = \varepsilon_{\infty} + \frac{C}{\left(\frac{T_1}{2}\right) \coth\left(\frac{T_1}{2T}\right) - T_0}, \quad (1)$$

where ε_{∞} is a temperature-independent background, T_1 is the characteristic quantum temperature, T_0 is a Curie–Weiss-like temperature, and the constant C sets the overall scale of the dielectric response. A low-temperature broad maximum in $\varepsilon'_c(T)$ may arise either from coupling of a soft optical phonon to the acoustic branch [22], as recently established in SrTiO₃ [23], or from coupling to defects [24,25]. In the present material, the relatively modest value of ε'_c , compared with classical large-permittivity quantum paraelectrics, does not favor a strong optical–acoustic phonon coupling scenario. In the related magnetoplumbite compound EuAl₁₂O₁₉, the

relevant polar phonon remains above ~ 3 THz [11], which also makes such a scenario less likely here. Below about 30 K, however, the data show a reproducible deviation from the pure Barrett form. In view of the previously established Nd³⁺ CEF scheme, we interpret this feature as the dielectric signature of thermal depopulation of the first excited CEF doublet. We therefore fit the permittivity using a sum of the Barrett background and an additional term associated with this excited-doublet contribution [26–31],

$$\varepsilon'_c(T) = \varepsilon'_{\text{Barrett}}(T) + \Delta\varepsilon'_{\text{CEF}}(T), \quad (2)$$

with

$$\Delta\varepsilon'_{\text{CEF}}(T) = \frac{A}{T} \text{sech}^2\left(\frac{\Delta}{2T}\right), \quad (3)$$

where Δ represents the energy separation of the first excited doublet expressed in kelvin, while A sets the magnitude of its dielectric contribution projected onto the measured axis, with $\Delta\varepsilon'_{\text{CEF}}(T) \rightarrow 0$ as $T \rightarrow 0$. A fit of the zero-field $\varepsilon'_c(T)$ in Fig. 1(a) using Eqs. (1)–(3) over the full measured range 0.3–285 K yields $\varepsilon_{\infty} = 18.44(1)$, $T_1 = 147.9(9)$ K, $T_0 = 5(1)$ K, $C = 141(2)$ K, $A = 2.80(5)$ K, and $\Delta = 25.9(3)$ K. Reducing the upper fitting limit to lower temperatures shifts the Barrett parameters somewhat—in particular, T_1 decreases by a few kelvin and T_0 becomes negative—but the fitted value of Δ remains robust against the choice of fitting window. The resulting gap, $\Delta = 25.9(3)$ K, agrees well with the lowest Nd³⁺ crystal-electric-field excitation previously estimated from point-charge calculations, $E_1 \simeq 2.17$ meV (≈ 25.2 K) [17]. This agreement supports direct assignment of the 25–30 K dielectric anomaly to thermal depopulation of the first excited CEF doublet. The dielectric response therefore suggests three superposed components: (i) a Barrett-like host background, (ii) an additional largely frequency-independent contribution on the 25–30 K scale associated with the first excited CEF doublet, and (iii) a low-temperature dispersive channel discussed below. The frequency-dependence of the real permittivity is illustrated in Figs. 1(b) and 1(c) up to 50 K in $\mu_0 H = 0$ and 9 T, respectively. At elevated temperatures, the permittivity is nearly frequency-independent. Below ~ 30 –35 K, however, the response develops a clear frequency-dependence. This dispersion is consistent with the

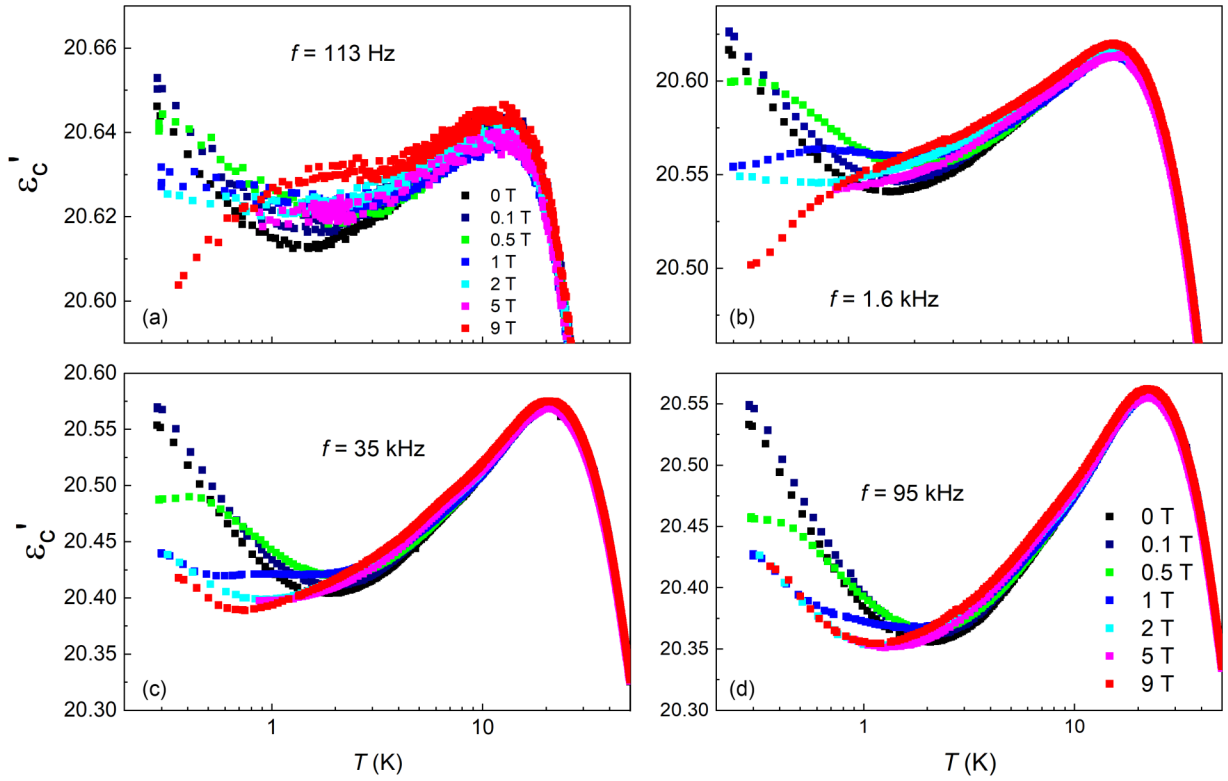


FIG. 2. Temperature dependence of $\varepsilon'_c(T)$ of $\text{NdMgAl}_{11}\text{O}_{19}$ in magnetic fields $\mu_0 H = 0\text{--}9\text{ T}$. (a) $f = 113\text{ Hz}$, (b) $f = 1.6\text{ kHz}$, (c) $f = 35\text{ kHz}$, and (d) $f = 95\text{ kHz}$.

type of disorder-related mechanism discussed previously for $\text{CeMgAl}_{11}\text{O}_{19}$, namely local inhomogeneity associated with random Mg/Al site mixing [10]. This dispersive contribution should be distinguished from the deviation of the zero-field temperature dependence from the pure Barrett law, which is captured here by the additional term associated with thermal depopulation of the first excited CEF doublet. The weak frequency-dependence of the 25–30 K anomaly position, together with the stability of the fitted gap against the fitting window, supports treating this feature separately from the broader dispersive response that develops below $\sim 30\text{--}35\text{ K}$. As in $\text{EuAl}_{12}\text{O}_{19}$, one may also consider a contribution from correlated antipolar dipolar dynamics [11]; however, in the present case the agreement between the fitted Δ and the previously inferred first excited CEF level supports assignment of the 25–30 K feature to the first excited-state CEF doublet, while the accompanying dispersion is consistent with disorder broadening of the response. In addition, the zero-field temperature dependence in Fig. 1(a) shows a further upturn of $\varepsilon'_c(T)$ below about 2 K, indicating another low-temperature contribution beyond both the broader dispersion observed below $\sim 30\text{ K}$ and the anomaly on the 25–30 K scale. A qualitatively similar low-temperature upturn has also been observed in $\text{CeMgAl}_{11}\text{O}_{19}$ and $\text{PrMgAl}_{11}\text{O}_{19}$, suggesting that this feature may reflect a broader aspect of the magnetoplumbite hexaaluminate family rather than a phenomenon unique to the Nd compound [9,10]. The onset of this upturn near 2 K is comparable to the energy scale set by the Curie–Weiss temperature $\Theta_{\text{CW}} \approx -0.5\text{ K}$ [17], suggesting that the development of antiferromagnetic

correlations among the Nd^{3+} moments plays a role. Cole–Cole analysis of $\varepsilon'_c(\omega, T)$ (see Supplemental Material [26]) shows that the relaxation frequency f_{rel} is essentially temperature independent from 0.3 to 10 K at zero field; neither an Arrhenius law nor a Vogel–Fulcher law fits the data, ruling out both thermally activated dynamics and relaxor-type freezing as the origin of the dispersion. The precise microscopic mechanism remains phenomenological at this stage. In this family, rare-earth ions are known to exhibit local off-centering within the oxygen bipyramidal site [17,19–21], which modifies the Nd–O–Nd superexchange geometry and generates a distribution of exchange interactions. Within this picture, the sub-2 K dielectric upturn and its associated dispersion arise from antiferromagnetically correlated Nd^{3+} clusters whose slow internal dynamics—governed by the distributed exchange landscape—couple to the polarizable host lattice and produce a frequency-dependent enhancement of ε'_c . The field evolution of the dielectric response is shown in Fig. 2, where $\varepsilon'_c(T)$ is plotted for $\mu_0 H = 0\text{--}9\text{ T}$ at representative frequencies. The broad feature near $\sim 25\text{--}30\text{ K}$ remains visible throughout the measured field range. By contrast, the low-temperature response becomes increasingly field and frequency-dependent. Below about 2 K, $\varepsilon'_c(T)$ varies substantially across the measured frequency window, with the differences becoming more pronounced at higher fields. For fields up to $\mu_0 H = 0.5\text{ T}$, the low-temperature permittivity shows the same qualitative trend at all measured frequencies, namely an upturn upon cooling. For fields of $\mu_0 H \geq 1\text{ T}$, this behavior becomes distinctly frequency-dependent. In

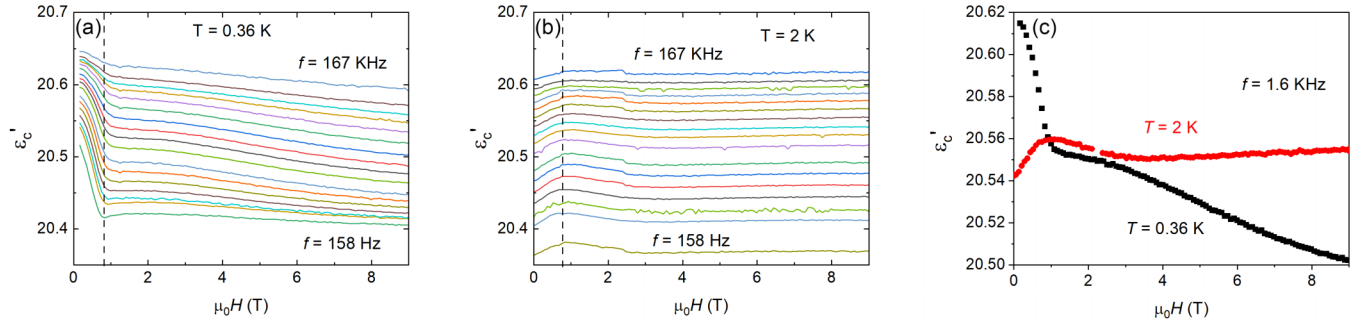


FIG. 3. Field dependence of the real permittivity $\epsilon'_c(H)$ of $\text{NdMgAl}_{11}\text{O}_{19}$. Panels (a) and (b) show measurements at several frequencies between 158 Hz and 167 kHz at (a) $T = 0.36$ K and (b) $T = 2$ K. Panel (c) compares $\epsilon'_c(H)$ measured at the selected frequency $f = 1.6$ kHz for $T = 0.36$ K and $T = 2$ K. The vertical dashed line marks the crossover field $\mu_0H_c \simeq 0.85$ T.

particular, at 9 T the low-temperature upturn is suppressed at lower frequencies [Figs. 2(a) and 2(b)], whereas it reappears at higher frequencies [Figs. 2(c) and 2(d)]. Further information is obtained from isothermal field sweeps. Figure 3 shows $\epsilon'_c(H)$ at $T \simeq 0.36$ K and 2 K for several frequencies between 158 Hz and 167 kHz. At both temperatures, the data show a change in field dependence near $\mu_0H_c \simeq 0.85$ T across the full measured frequency window. At 0.36 K this low-field scale appears as a pronounced knee separating a steep initial suppression of ϵ'_c from a much weaker high-field evolution. At 2 K, the response over the same field range becomes broader and changes sign, with an initial low-field enhancement followed by flattening at higher field. In addition to the primary crossover near μ_0H_c , a weaker deviation from the smooth high-field trend is visible at both temperatures just below ~ 3 T, appearing as a small kink or shoulder in the field dependence.

Discussion. The results above reveal two characteristic low-energy scales superposed on a Barrett-like host background. We now discuss their interpretation, beginning with the 25–30 K anomaly and then turning to the low-temperature field-dependent response. The close agreement between the fitted gap $\Delta = 25.9(3)$ K and the first excited Nd^{3+} CEF doublet at $E_1 \simeq 2.17$ meV (≈ 25.2 K) from point-charge analysis [17] strongly supports assignment of the 25–30 K dielectric anomaly to thermal depopulation of this doublet. Such CEF–dielectric coupling is consistent with the broader observation that rare-earth CEF excitations can leave clear signatures in optical and magnetodielectric response through coupling to phonons and other low-energy modes [6–8]. The Nd^{3+} ground-state Kramers doublet is strongly anisotropic ($g_c \simeq 3.75\text{--}4.46$, $g_{ab} \simeq 1.4\text{--}1.5$; Table S1 within Supplemental Material [26]), with Zeeman splitting $\Delta E_Z \simeq 22.7\text{--}27.0$ K at $\mu_0H = 9$ T (Table S2 within Supplemental Material [26]); nevertheless, the anomaly position is field-independent because both Kramers doublets split symmetrically about their respective zero-field center-of-mass energies, leaving the average inter-manifold gap near E_1 [32–34].

The low-temperature dielectric response involves a competition among three energy scales whose relative importance changes with temperature: (i) antiferromagnetic exchange correlations among the Nd^{3+} moments, enhanced by the distribution of exchange paths arising from rare-earth

off-centering; (ii) Zeeman splitting of the ground-state Kramers doublet; and (iii) the thermal energy $k_B T$. At $T = 0.36$ K, the system lies well below $|\Theta_{\text{CW}}| \approx 0.5$ K ($\Theta_{\text{CW}} \approx -0.5$ K [17]), placing it within the regime of frustrated antiferromagnetic correlations on the triangular Nd^{3+} sublattice. In this regime, thermal broadening alone cannot account for the observed crossover: the Cole–Cole broadening parameter α extracted from the zero-field permittivity remains nearly temperature independent ($\alpha \simeq 0.60\text{--}0.65$ between 0.3 and 10 K; see Supplemental Material [26]), whereas a static disorder distribution of activation energies would yield $\alpha \rightarrow 1$ as $T \rightarrow 0$ [31]. A weakly temperature-dependent α of comparable magnitude is instead characteristic of broadly distributed cluster or glassy dynamics in correlated magnets [11,31]. The pronounced suppression of ϵ'_c at 0.36 K under field application up to μ_0H_c [Fig. 3(a)] then provides direct evidence that antiferromagnetic correlations contribute to the low-field permittivity, since static structural disorder would not be appreciably suppressed by such modest fields. We interpret this contribution as arising from antiferromagnetically correlated Nd^{3+} clusters—facilitated by the distribution of exchange interactions because of rare-earth off-centering [17,19–21]—whose internal spin dynamics modulate the local dielectric response through exchange-mediated coupling to the polarizable host lattice [8,11]. The frequency-dependence of this low-temperature contribution provides a key constraint on its microscopic origin. A static spin–phonon-coupling mechanism, in which antiferromagnetic correlations soften an optical phonon and thereby enhance ϵ'_c via the Lyddane–Sachs–Teller relation, would produce an essentially frequency-independent shift across our measurement window, since the relevant transverse-optic mode in the magnetoplumbite hexaaluminates lies above ~ 3 THz [11]—more than seven decades above our highest measurement frequency. The strong dispersion observed below 2 K therefore rules out a purely magnetoelastic (soft-mode) origin and instead points to slow relaxational dynamics of the correlated-cluster manifold, whose characteristic frequencies fall within the Hz–kHz range probed here. The Zeeman splitting at the crossover, $\Delta E_Z \approx 2.2$ K (Table S2 within Supplemental Material [26]; $g_{\text{eff}} = 3.86(5)$ from Schottky analysis [17]), is approximately four times $|\Theta_{\text{CW}}|$, placing the crossover quantitatively at the boundary where Zeeman splitting overcomes the

TABLE I. Cole–Cole parameters below 2 K at $\mu_0 H = 0$ and 9 T. $f_{\text{rel}} = (2\pi\tau)^{-1}$; α is the broadening exponent. Full analysis in Supplemental Material [26].

T (K)	$\mu_0 H = 0$ T		$\mu_0 H = 9$ T	
	f_{rel} (kHz)	α	f_{rel} (kHz)	α
0.35	22.8	0.63(3)	0.82	0.42(2)
0.45	20.4	0.64(2)	1.15	0.48(2)
0.67	17.3	0.64(2)	4.24	0.55(2)
0.95	20.0	0.64(2)	8.85	0.56(2)

antiferromagnetic correlations [1,2], and we accordingly identify $\mu_0 H_c$ as the field at which Zeeman splitting overcomes the antiferromagnetic correlations and drives the system out of the correlated regime. By contrast, at $T \simeq 2$ K the isothermal magnetization follows a Brillouin-function form for an effective spin- $\frac{1}{2}$ doublet [17], indicating that antiferromagnetic correlations already play a reduced role and the response is closer to a single-ion limit. At this temperature, thermal energy enters as the third competing scale: the field response becomes broader, the low-field response changes sign, and the crossover near $\mu_0 H_c$ loses the sharp knee seen at 0.36 K. The smooth evolution from sharp at 0.36 K to broad at 2 K thus reflects the progressive dilution of correlation effects by thermal fluctuations. In the high-field regime ($\mu_0 H \gtrsim \mu_0 H_c$), the dielectric response is governed by the Zeeman-split ground-state Kramers doublet. Simple population redistribution between the two field-split states cannot drive the slow, frequency-dependent dispersion observed here, since the static two-level susceptibility is essentially frequency-independent over the Hz–kHz range [31]. Instead, the field dependence reflects the dynamics of inter-state transitions—moment-flipping events—between the Zeeman-split branches, whose characteristic rate is Boltzmann-suppressed as $\exp(-\Delta E_Z/k_B T)$ and therefore decreases rapidly with increasing field. This is directly observed in the Cole–Cole analysis (Table I): at $\mu_0 H = 9$ T, f_{rel} drops by more than an order of magnitude below 2 K while α decreases from 0.56 to 0.49, in contrast to the nearly temperature-independent $f_{\text{rel}} \approx 17\text{--}23$ kHz and $\alpha \approx 0.64$ at zero field; full Cole–Cole analysis is given in Supplemental Material [26]. The simultaneous slowing and partial Debye-like sharpening of the relaxation is consistent with a progressively better-defined single-ion moment-flipping process. The primary crossover near $\mu_0 H_c$ does not exhaust the full high-field response. In Fig. 3(c), a weaker shoulder-like deviation is visible at both temperatures just below ~ 3 T. A natural interpretation is that $\mu_0 H_c \simeq 0.85$ T marks the principal exit from the correlated regime, whereas the feature near 2–3 T reflects a more gradual approach to a fully Zeeman-split single-ion limit in which the field-induced splitting exceeds the residual broadening from local CEF disorder and weak effective exchange [26,35]. These are not two distinct thermodynamic boundaries but rather two stages of the same evolution of a broadened low-energy manifold under field. Structural and spectroscopic

considerations support such a broadened picture. Local structural disorder has been reported for $\text{NdMgAl}_{11}\text{O}_{19}$ and related magnetoplumbite hexaaluminates from SCXRD studies, which reveal local deviations from an ideal single-site rare-earth environment [17,19,20,36,37]. Rare-earth CEF spectra are highly sensitive to such local variations: distinct local charge surroundings can produce additional CEF excitations [38], while smooth environmental disorder broadens the spectrum into a wide envelope [39]. Exchange and fluctuation effects add further linewidth [40]. In $\text{NdMgAl}_{11}\text{O}_{19}$, the relevant low-energy limit is therefore a ground-state doublet manifold broadened by distributed local CEF parameters, strain fields, and exchange couplings arising from rare-earth off-centering, on top of which the antiferromagnetic correlations develop at the lowest temperatures. This behavior differs from that of the isostructural Kramers compound $\text{CeMgAl}_{11}\text{O}_{19}$, where the dielectric anomaly appears near $T^* \approx 3.6$ K and was discussed in terms of higher-order virtual CEF processes rather than direct thermal depopulation [10]. The contrast is sharper still for the non-Kramers compound $\text{PrMgAl}_{11}\text{O}_{19}$, where off-centering of the Pr ion lifts the non-Kramers degeneracy already in zero field, producing a distribution of low-energy scales and a broader dielectric and thermodynamic response [9,19]. In $\text{NdMgAl}_{11}\text{O}_{19}$, the ground-state doublet remains Kramers protected in zero field, but its experimental manifestation is broadened by disorder and, at the lowest temperatures, by antiferromagnetic correlations. Comparison with $\text{EuAl}_{12}\text{O}_{19}$, where the dielectric response is dominated by the frustrated AlO_5 dipolar subsystem with no rare-earth CEF control [11], completes the sequence: $\text{EuAl}_{12}\text{O}_{19}$ represents the dipolar host, $\text{CeMgAl}_{11}\text{O}_{19}$ shows indirect CEF control, $\text{PrMgAl}_{11}\text{O}_{19}$ shows a distributed non-Kramers response, and $\text{NdMgAl}_{11}\text{O}_{19}$ provides a Kramers reference system in which both excited-doublet depopulation and a field-driven crossover from correlated to single-ion behavior are reflected in the permittivity.

Conclusions. Dielectric spectroscopy of single-crystalline $\text{NdMgAl}_{11}\text{O}_{19}$ reveals three superposed contributions to the permittivity along the c axis. At higher temperatures, $\epsilon'_c(T)$ follows a Barrett-like quantum-paraelectric background. Below ~ 30 K, a robust additional contribution with $\Delta = 25.9(3)$ K is assigned to thermal depopulation of the first excited Nd^{3+} CEF doublet [17]. At the lowest temperatures, the response becomes strongly frequency and magnetic-field dependent. The crossover at $\mu_0 H_c \simeq 0.85$ T reflects a competition between antiferromagnetic correlations—whose relaxational, cluster-like character is evidenced by the strong frequency-dependence and the nearly constant Cole–Cole broadening parameter (Table I)—and Zeeman splitting of the ground-state Kramers doublet. $\text{NdMgAl}_{11}\text{O}_{19}$ thus serves as a Kramers reference system for separating excited-state CEF signatures from those of the field-tuned, correlation-dominated ground-state sector in centrosymmetric frustrated rare-earth hexaaluminates.

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Data availability. The data that support the findings of this Letter are not publicly available. The data are available from the authors upon reasonable request.

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