Search for the magnetic monopole at a magnetoelectric surface

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Abstract The elusiveness of magnetic monopoles, which are expected in classical electrodynamics because of the duality symmetry between electricity and magnetism, has intrigued physicists for centuries. Their relevance was particularly emphasized by Dirac, who introduced a description allowing monopoles to remain consistent with the known zero divergence of magnetic fields, and showed that their existence would explain the observed quantization of electric charge in the universe. The quest for a magnetic monopole therefore remains an active research area today, ranging from searches using sensitive cosmic-ray detectors to attempts to generate monopoles in collider experiments; for a review see Ref. 2.

While the existence of true magnetic monopoles has not yet been verified, a number of condensed-matter systems have been shown to provide intriguing analogues. Perhaps the most popular are the pyrochlore-structure “spin-ice” materials of which the prototype is dysprosium titanate, Dy2Ti2O7. In these materials, magnetic excitation of the frustrated antiferromagnetic “two-in, two-out” tetrahedral spin ordering leads to two locally divergent magnetizations of opposite sign – one tetrahedron has three spins pointing inward and one pointing outward, and vice versa – connected by the analogue of a Dirac string. Also of interest are the so-called linear magnetoelectric materials, magnetic insulators in which an applied electric field induces a magnetization and vice versa. Here, it has been shown theoretically that when an electric charge is introduced into a diagonal magnetoelectric (in which the induced magnetization is parallel to the electric field), the divergent electric field of the charge induces a monopole-like magnetization around the electric charge. Similarly, it has been argued that a charge above a topological insulator/ferromagnetic heterostructure should lead to a magnetic monopolar field due to the quantized Chern-Simons magnetoelectric response of topological insulators with broken time-reversal symmetry. While the magnetoelectric response of such a system can in principle be sizable, its detection is challenging because of the practical difficulty in achieving insulating bulk behavior in topological insulators, as well as the need to incorporate a separate time-reversal symmetry breaking component.

Here we show that conventional linear magnetoelectric materials, of which Cr2O3 is the prototype, can generate an external monopolar magnetic field when an electric charge is placed above any flat sample surface. In linear magne-
electrics, an applied magnetic field \( \mathbf{H} \) induces an electric polarization \( \mathbf{P} \) and an applied electric field \( \mathbf{E} \) induces a magnetization \( \mathbf{M} \) according to

\[
P = \varepsilon \mathbf{H} \\
\mu_0 \mathbf{M} = \sigma^T \mathbf{E}.
\]

Here \( \mu_0 \) is the permeability of free space and \( \sigma \) is the magnetoelectric tensor in SI units, which is allowed to be non-zero in materials that break both time-reversal and space-inversion symmetry, and which has non-zero components determined by the detailed crystalline and magnetic symmetry. We show theoretically that, in cases for which \( \sigma \) has a non-zero diagonal component, a surface charge \( q \) generates a sub-surface image monopole \( m \). This leads in turn to a divergent magnetic field above the sample surface, which we detect using low energy muon spin rotation spectroscopy.

II. CALCULATION OF THE FIELDS INDUCED BY A CHARGE ON THE SURFACE OF A MAGNETOELECTRIC MATERIAL

We consider the geometry shown in Fig. 1, in which a point electric charge \( q \) is placed in the vacuum region a small distance \( r_0 = (0, 0, z_0) \) away from the planar surface of a semi-infinite slab of a uniaxial magnetoelectric material.

A. Magnetoelectrostatics

We solve the classical Maxwell equations for a static system in which the electromagnetic fields are given by Gauss’ laws

\[
\mathbf{D} = \varepsilon \mathbf{E} + \sigma^T \mathbf{H} \\
\mathbf{B} = \mu \mathbf{H} + \sigma \mathbf{E},
\]

where \( \sigma^T \) is the linear magnetoelectric susceptibility tensor and \( \sigma^T \) its transpose. This expanded formulation must be used in the Maxwell equations \( \mathbf{D} \) and \( \mathbf{B} \) to calculate the electromagnetic fields in a magnetoelectric material. In addition, the system needs to satisfy the electrostatic boundary conditions for interfaces at all times:

\[
\mathbf{D} \cdot \mathbf{n} = \text{constant} \\
\mathbf{B} \cdot \mathbf{n} = \text{constant} \\
\mathbf{E} \cdot \mathbf{t} = \text{constant} \\
\mathbf{H} \cdot \mathbf{t} = \text{constant},
\]

where \( \mathbf{n} \) is the surface normal and \( \mathbf{t} \) the surface tangent.

Since we look at the static limit, it is helpful to use the electrostatic and magnetostatic potentials, \( \phi_e \) and \( \phi_m \), which are related to the electric and magnetic fields by

\[
\mathbf{E} = -\nabla \phi_e \\
\mathbf{H} = -\nabla \phi_m.
\]

B. Solution for an isotropic magnetoelectric

First, we present the solution of the field equations for a charge above an isotropic magnetoelectric in which \( \sigma = \alpha \mathbf{I} \) (\( \mathbf{I} \) is the unit matrix). Even though there are five magnetic point groups permitting such behavior, no material with such a magnetoelectric response has yet been identified experimentally. Nevertheless, the behavior is of academic interest, since it has the symmetry of the so-called Chern-Simons magnetoelectric response of topological insulators\(^{15}\). In addition, the solution is obtained straightforwardly using the well-established method of mirror charges, and already provides insight into the full problem that we address in the next section. Placing mirror charges inside, \( \text{in} \), and outside, \( \text{out} \), of the magnetoelectric we obtain the ansatz for the electric potential, \( \phi_e \):

\[
\phi_e^{\text{out}}(r) = \frac{q}{4\pi \varepsilon_0} \frac{1}{|r - r_0|} + \frac{q'}{|r - r_1|} \tag{13}
\]

\[
\phi_e^{\text{in}}(r) = \frac{q''}{|r - r_0|}, \tag{14}
\]

where \( q \) is the real charge, and \( q' \) and \( q'' \) are electric image charges at positions \( r_0 = (0, 0, z_0) \), \( r_1 = (0, 0, -z_0) \). We enforce continuous normal components of the displacement field and magnetic flux density at the interface as well as continuous tangential components of the electric and magnetic fields. To satisfy the magnetic boundary conditions, we use the following ansatz for the magnetic potential:

\[
\phi_m^{\text{out}}(r) = \frac{m'}{|r - r_1|} \tag{15}
\]

\[
\phi_m^{\text{in}}(r) = \frac{m''}{|r - r_0|}, \tag{16}
\]

where \( m' \) and \( m'' \) are effective magnetic image monopoles at positions \( r_0 = (0, 0, z_0) \), \( r_1 = (0, 0, -z_0) \). We solve this system of equations, as shown in detail in Appendix \( A \) to obtain the following expression for the magnetic flux density outside of the material:

\[
\mathbf{B}(r) = -\frac{\mu_0}{4\pi (\mu + \mu_0)(\varepsilon + \varepsilon_0) - \alpha^2} \frac{q|\alpha|}{|r - r_1|^3} \tag{17}
\]

The resulting \( \mathbf{E} \) and \( \mathbf{B} \) fields both inside and outside of the magnetoelectric slab, using literature values for the response parameters of \( \text{Cr}_2\text{O}_3 \) (Table 1) averaged to mimic an isotropic material, are sketched in Figs. 2(a) and b).

The electric field outside the slab is similar to that of the original isolated point charge, with deviations in the region.
close to the interface due to the dielectric screening of the field within the slab. The electric field within the slab is a divergent point charge field with the charge outside the slab as its origin, and its magnitude screened by the static dielectric constant of the material. Since the magnetic flux density within the material is induced by the electric field through the magnetoelectric effect, the field lines within the slab diverge identically to those of the electric field. Outside of the slab, the magnetic field is particularly interesting as it is perfectly divergent, with its source being an image monopole that is the same distance below the surface as the point charge is above it. A positive charge with the magnitude of an electronic charge induces an image monopole $m^\perp = 3.63 \times 10^{-16}$ A m in a material with these response parameters. This converts to a magnetic $B$-field of the order of $5 \mu T$ by and measured at the site of a single electronic point charge placed a distance of $2 \text{ nm}$ above the interface. Note that a positive charge on a material with a positive magnetoelectric tensor induces a negative magnetic field outside the material, and that changing the sign of one of the surface charge or the magnetoelectric tensor changes the sign of the field. As a result, opposite magnetoelectric domains produce fields of opposite sign for the same sign of charge.

### C. Solution for a uniaxial magnetoelectric

Next we analyze the realistic case of the response of an uniaxial anisotropic magnetoelectric material [19]. Specifically, we take the case of the prototypical magnetoelectric, Cr$_2$O$_3$, and treat its full uniaxial response. We orient the high-symmetry axis along the $z$ axis, so that the magnetoelectric, dielectric and magnetic susceptibility tensors are as follows:

$$\bar{\alpha} = \begin{bmatrix} \alpha_\parallel & 0 & 0 \\ 0 & \alpha_\perp & 0 \\ 0 & 0 & \alpha_\perp \end{bmatrix}, \ \bar{\varepsilon} = \begin{bmatrix} \varepsilon_\parallel & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_\perp \end{bmatrix}, \ \bar{\mu} = \begin{bmatrix} \mu_\perp & 0 & 0 \\ 0 & \mu_\parallel & 0 \\ 0 & 0 & \mu_\parallel \end{bmatrix}$$

Aligning the $n = (0,0,z)$ axis of the magnetoelectric perpendicular to the surface plane, the field equations inside the magnetoelectric become

$$\nabla \cdot D = (\varepsilon_\perp \nabla_\perp + \varepsilon_\parallel \nabla_\parallel) E + (\alpha_\perp \nabla_\perp + \alpha_\parallel \nabla_\parallel) H = 0$$
$$\nabla \cdot B = (\mu_\perp \nabla_\perp + \mu_\parallel \nabla_\parallel) H + (\alpha_\perp \nabla_\perp + \alpha_\parallel \nabla_\parallel) E = 0,$$

where $D = \varepsilon_0 E$ and $E$ is the electric field. We solve this system of equations by Fourier transformation in the two-dimensional coordinate space perpendicular to the interface, and then solving separately for the two half spaces with the boundary conditions stated previously in section II A. We obtain the following expressions for the potentials $\phi_m$ and $\phi_e$ (for details see the Appendix):

$$\phi_m^\text{in} = \frac{c_{e1}^\text{in}}{\sqrt{R^2 + |\xi - z_0|^2}} + \frac{c_{e2}^\text{in}}{\sqrt{R^2 + |\xi - z_0|^2}}$$
$$\phi_e^\text{in} = \frac{1}{4\pi\varepsilon_0} \frac{q}{\sqrt{R^2 + |z - z_0|^2}} + \frac{q_e^\text{out}}{\sqrt{R^2 + |z - z_0|^2}}$$
$$\phi_m^\text{out} = \frac{c_{b1}^\text{in}}{\sqrt{R^2 + |\xi - z_0|^2}} + \frac{c_{b2}^\text{in}}{\sqrt{R^2 + |\xi - z_0|^2}}$$
$$\phi_e^\text{out} = \frac{c_{b1}^\text{out}}{\sqrt{R^2 + |\xi - z_0|^2}} + \frac{c_{b2}^\text{out}}{\sqrt{R^2 + |\xi - z_0|^2}}$$

with $R = \sqrt{x^2 + y^2}$, $\xi = \sqrt{\frac{y^2 + d^2}{2}}$ is determined by the

![Figure 2](image-url)
The values of the parameters for the case of Cr$_2$O$_3$ (obtained using the susceptibilities from table I) are given in table II. Eqn. 23 leads us immediately to the central result of our calculation:

$$B(r) = \mu_0 e^{out} \frac{r - r_1}{|r - r_1|^3}.$$  

(24)

Here $r = (x, y, z)$ and $r_1 = (0, 0, -z_0)$. We plot the magnetic field in Fig. 3 for the parameters of Cr$_2$O$_3$. The monopolar nature above the surface is clear, while the behavior beneath the surface is more complicated than in the isotropic case.

![Graph showing magnetic field $B$ along $(0,0,z)$ induced by an electronic charge, $q = +|e|$, 2 nm above the surface at $z = 0$ of the magnetoelectric slab. Positive $z$ values are above the sample surface. The field is decomposed into contributions from the isotropic and anisotropic components of the magnetoelectric tensor. We see that the monopolar field outside the sample is determined entirely by the isotropic component of the magnetoelectric response.](image)

FIG. 3. Magnetic field $B$ along $(0,0,z)$ induced by an electronic charge, $q = +|e|$, 2 nm above the surface (at $z = 0$) of the magnetoelectric slab. Positive $z$ values are above the sample surface. The field is decomposed into contributions from the isotropic and anisotropic components of the magnetoelectric tensor. We see that the monopolar field outside the sample is determined entirely by the isotropic component of the magnetoelectric response.

Note that the electric field (not shown) is indistinguishable from that obtained for the isotropic case because it is dominated by the dielectric response, which is almost isotropic. The additional electric polarization that is induced by the magnetoelectric response is negligible compared to the direct dielectric response. We emphasize again that, due to the transformation properties of the magnetoelectric tensor, the sign of the magnetic image charges, and the corresponding induced $B$ field, will be opposite in the two different AFM domains of Cr$_2$O$_3$.

### D. Dependence of the monopolar field strength on the magnetoelectric anisotropy

We saw in the previous two sections that the induced monopolar field depends on both the magnitude of the magnetoelectric response and its anisotropy, that is the relative magnitudes of $\alpha_\parallel$ and $\alpha_\perp$. In Appendix C we give a detailed analysis of the effect of anisotropy, the main results of which we present here. We write the magnetoelectric tensor as a sum of isotropic (proportional to the sum of $\alpha_\parallel$ and $\alpha_\perp$) and anisotropic (proportional to the difference between $\alpha_\parallel$ and $\alpha_\perp$) contributions:

$$\bar{\alpha} = \frac{1}{2} (\alpha_\parallel + \alpha_\perp) \mathbb{I} + \frac{1}{2} (\alpha_\parallel - \alpha_\perp) \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{bmatrix},$$  

(25)

and show the field contributions from the two components for the case of Cr$_2$O$_3$ in Fig. 3 calculated assuming that the anisotropies in $\varepsilon$ and $\mu$ are small.

We see that, while both isotropic and anisotropic components of the magnetoelectric tensor contribute to the field within the slab, only the isotropic component is relevant for the field outside the magnetoelectric; in fact for the case of exactly isotropic $\bar{\tau}$ and $\bar{\mu}$ tensors the field outside the slab is given by the result that we derived for the fully isotropic case, Eqn. 17. This is consistent with the symmetry of the vacuum, in which a hypothetical magnetic charge would induce a purely monopolar magnetic field. Anisotropies in the $\bar{\tau}$ and $\bar{\mu}$ tensors modify the magnitude of $B(r)$ slightly from that of Eqn. 17 (for the case of Cr$_2$O$_3$ using the values from table I) we find a difference of 0.05% between the exact solution and that for averaged isotropic $\bar{\tau}$ and $\bar{\mu}$, but do not change its monopolar form.

This feature makes it particularly straightforward to predict the temperature dependence of the monopolar field. The highly temperature dependent magnetoelectric response in Cr$_2$O$_3$ is reproduced in Fig. 4. While the in-plane magnetoelectric response, $\alpha_\parallel$, shows the usual Brillouin-function form below the Néel temperature (orange triangles in Fig. 4), the spin-fluctuation mechanism responsible for the out-of-plane response, $\alpha_\perp$, results in a strong temperature dependence (green squares in Fig. 4), with $\alpha_\perp$ even changing sign at low...
temperature. Since the strength of the induced monopole is proportional to the isotropic component, \( \frac{1}{2}(\alpha_\parallel + \alpha_\perp) \), shown as the red line in Fig. 4, the corresponding induced monopolar field must have the same temperature dependence. We see that the induced monopolar field should increase with increasing temperature, reaching a maximum at around 280 K, before decreasing and vanishing at the Néel temperature at \(~ 310\) K.

### III. EXPERIMENTAL SEARCH FOR THE MAGNETIC MONOPOLE USING LOW ENERGY MUON SPIN ROTATION (LE-\(\mu\)SR)

Next we describe our experimental search for the magnetic monopolar field using low energy muon spin rotation (LE-\(\mu\)SR).

#### A. Experimental setup

In the LE-\(\mu\)SR method, fully polarized muons are implanted into a sample and the local magnetic field at the muon stopping site is measured by monitoring the evolution of the muon spin polarization. This is achieved via the anisotropic beta decay positron which is emitted preferentially in the direction of the muon's spin at the time of decay. Using appropriately positioned detectors one can measure the asymmetry, \( A(t) \), of the beta decay along the initial polarization direction. \( A(t) \) is proportional to the time evolution of the spin polarization of the ensemble of implanted spin probes.\(^{25}\)

Conventional \(\mu\)SR experiments use so-called surface muons with an implantation energy of 4.1 MeV, resulting in a stopping range in typical density solids of from 0.1 mm to 1 mm below the surface. As a result, their application is limited to studies of bulk properties and they cannot provide depth-resolved information or study extremely thin film samples. In contrast, depth-resolved \(\mu\)SR measurements can be performed at the low-energy muon (LEM) spectrometer using muons with tunable kinetic energies in the 1 keV to 30 keV range, corresponding to implantation depths of 10 nm to 200 nm. We take advantage of this capability here.

Our measurement is designed in the following way: We use a 500 nm thick Cr\(_2\)O\(_3\) film grown in the (001) direction, which is coated by an insulating stopping layer, in this case solid nitrogen, N\(_2\). The muons (which carry a positive electronic charge +e) are implanted at different depths in the N\(_2\) layer. The electric field of the muon should penetrate into the Cr\(_2\)O\(_3\) layer and induce both electric and magnetic responses, with the magnetic response being the monopolar field described above in this paper. The muon itself then acts as the magnetic probe to measure the induced magnetic field. The full experimental setup is sketched in Fig. 5.

The Cr\(_2\)O\(_3\) films used here were grown by reactive rf sputtering on (0001) Al\(_2\)O\(_3\) substrates using a metal Cr target in an Ar + O\(_2\) atmosphere (base pressure < 1 \(\times\) 10\(^{-6}\) Pa) at a substrate temperature of 773 K. Bottom Pt electrodes with thicknesses of 25 nm were sputtered on Al\(_2\)O\(_3\) substrates and Cr\(_2\)O\(_3\) films using shadow masking. Prior to our measurements, the Cr\(_2\)O\(_3\) layer was prepared in a single domain state using magnetoelastic annealing. This was achieved by cooling the sample from 320 K through the Néel temperature to 20 K in a positive magnetic field of 0.3 T and a positive electric field larger than 1 kV/cm, both applied along the surface normal. Since \( E \) and \( H \) are parallel, such an anneal yields a single magnetoelastic domain with positive magnetoelastic tensor \(\alpha_{\text{avg}}\).

We then deposited a 150 nm layer of solid nitrogen on top of the Cr\(_2\)O\(_3\) film to provide an insulating muon stopping region above the surface of the magnetoelastic. The N\(_2\) deposition and all subsequent measurements were performed at 20K to maintain the N\(_2\) in the solid state. Muons were then implanted into this bilayer structure with different incident muon kinetic energies, in the presence of a small bias.

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**FIG. 4.** Measured temperature dependence of the parallel (\(\alpha_\parallel\), green squares) and perpendicular (\(\alpha_\perp\), orange triangles) magnetoelectric response in Cr\(_2\)O\(_3\). The red circles show the average, \(\frac{1}{2}(\alpha_\parallel + \alpha_\perp)\). Data taken from Ref.17.

**FIG. 5.** Sketch of the LE-\(\mu\)SR setup used for this experiment. Muons with a kinetic energy of 12.7 keV enter the sample region with nearly 100% spin polarization (red arrow). The energy of the muons impinging on the sample can be tuned by choosing the appropriate potential at the sample plate. The sample was cooled in a positive poling field of \(B_{\text{pol}} = 0.3\) T and a positive electric field of \(E > 1\) kV/cm to ensure a positive \(\alpha\). The measurements at low temperature were performed in \(B_{\text{meas}} = \pm 10\) mT.
field, $B_{\text{meas}} = \pm 10$ mT. The fraction of muons that do not capture an electron to form the neutral hydrogen-like muonium state is about 40% in the N$_2$ film [29]. The muonium response occurs at a completely different resonance frequency and so is easily subtracted from the measurement. The bias field is used to increase the accuracy of the measurement, but is too small to reorient the antiferromagnetic domain and so does not change the sign of the magnetoelectric tensor [30]. We performed independent second harmonic generation domain imaging experiments [31] and verified that the domain structure is stable up to fields of 5.8 T (at which a spin-flop occurs).

To check that our experiment has the required accuracy to detect the proposed monopolar field, we first calculated the muon stopping profiles for different muon implantation energies and assuming an N$_2$ thickness of 150 nm using the Monte Carlo program TRIM.SP. This program treats the positive muon as a light proton and has been shown to be accurate for low-energy muons [29]. Our calculated muon fractions as a function of implantation depth are shown for various implantation energies in the inset of Fig. 6. Our calculated LE-µSR fractional asymmetry as a function of implantation energy is shown by the black line in the main panel; this decreases for increasing energies as the muons start to reach the magnetic Cr$_2$O$_3$ layer where they quickly lose their polarization due to the strong internal magnetic fields. Also plotted in the main panel is our measured LE-µSR asymmetry measured in a transverse magnetic field of $\pm 10$ mT. The agreement in trend between the results of the TRIM.SP calculations and the measured values indicate that the nitrogen layer is indeed 150 nm thick.

Combining our calculated monopolar field strengths from Fig. 5 with the calculated and experimentally verified muon stopping distributions of Fig. 3, we show in Fig. 7 the calculated fraction of muons that experience fields between zero and 0.2 mT for implantation energies of 4, 7, 10 and 12 keV. We see that, for example for an energy of 12 keV, approximately 50% of the muons experience a magnetic field larger than 0.08 mT, which can be measured routinely using low-energy µSR. Additionally, the corresponding full width at half maximum of the monopole field distribution is of the same order (0.05 mT), which should lead to an experimentally measurable depolarization of the precessing muon spin ensemble.

### B. Results

In Fig. 8(a), we show the measured internal fields at the muon sites as a function of the muon implantation energy, with higher implantation energies corresponding to smaller average distances to the Cr$_2$O$_3$ surface. The upper panel (blue circles) shows the results obtained in small positive bias field (along +c), and the lower panel (red circles) those obtained in a small negative bias field. The internal field shown in Fig. 8 is the sum of the bias field plus any internal field. We see that in both cases the muon experiences a local magnetic field that varies monotonically with its distance from the surface. Note that only the muons stopping in the nitrogen overlayer contribute to the signal as the muons stopping in Cr$_2$O$_3$ quickly depolarize. The LE-µSR raw-data for an example point is shown in Appendix D.

If the only contribution to the internal field at the muon site were the monopolar field from the magnetoelectric response, we would expect the shifts in both cases to be in the same direction, since both sets of measurements are performed on the same magnetoelectric domain. It is known, however, that Cr$_2$O$_3$ surfaces have a surface magnetization [32–34], which has been shown to be susceptible to small magnetic fields in thin film samples [35]. To remove the contribution from the surface magnetic dipole, which we expect to switch with the bias measurement field, we therefore sum the local internal values obtained in positive and negative bias, and present $0.5(B_+ + B_-)$
as a function of muon energy in Fig. 8(b). The base level bias corresponds to the switching precision of the small magnetic bias field. We obtain an internal field shift that is consistent with the expected behavior of the induced magnetic monopole: The maximum value close to the surface is of the same order of magnitude (tens of µT) as the calculated value, the sign is as expected for the prepared magnetoelectric domain, and it decays with distance from the interface. While the size of the error bars prohibits extraction of the exact functional form, the decay is consistent with quadratic behaviour.

Another measure for the local fields near the Cr$_2$O$_3$ interface is the depolarization rate of the muon spin ensemble. It is directly proportional to the width of the distribution of fields sensed by the muons. We extracted this width by assuming a Lorentzian distribution of fields $P_L(B)$ when fitting the experimental data. The results are shown in Fig. 8(c). Encouragingly, the observed increase of the full width at half maximum is very close to that expected for the proposed monopole effect that can be extracted from Fig. 7, and clearly increases for both bias field orientations on approaching the surface.

IV. DISCUSSION AND OTHER EXPERIMENTAL TECHNIQUES

The small field shift in our LE-µSR measurements, combined with the increased width of the field distribution towards the interface present a first hint that a monopole is indeed induced by an electric charge at a magnetoelectric surface. In this final section we discuss studies that we have attempted using other techniques, as well as additional possible future routes for confirmation of the monopole’s existence.

A first step would be to perform temperature-dependent measurements using the LE-µSR technique described above. We showed in section II D the temperature dependence of the average magnetoelectric response, which in turn determines the strength of the monopolar field. A measured increase in field strength on warming with a maximum at around 280 K would be a strong indication that the origin of the field is the magnetoelectric response of the sample. For such a study, a different stopping layer would be needed because nitrogen would not be solid.

A. Magnetic force microscopy

In addition to the muon experiments we performed magnetic force microscopy (MFM) on a cut and etch-polished commercial c-oriented Cr$_2$O$_3$ crystal of $d = 150 \ \mu$m thickness grown by the Verneuil method (Kristallhandel Kelpin). The magnetic tip of an atomic force microscope acted as a charge monopole by applying a voltage $U$ of 20 V between the tip and the copper back electrode of the sample. At the same time, the magnetization of the tip served as the detector for the induced monopolar magnetic field. The goal of the experiment was to exploit the different sign of $\alpha$ for the two antiferromagnetic domains and measure a change of sign in the response when the tip moves across a domain boundary, as sketched in Fig. 9.
In addition we aimed to vary the tip-surface distance to verify the characteristic $r^2$ dependence of a monopolar field. From $U$, $d$ and $\alpha$ we estimated the monopolar field at the position of the tip to be on the order of 1 $\mu T$, which is detectable as a change of the mechanical deformation of the magnetized tip.

In the first step, we determined the distribution of antiferromagnetic domains in our Cr$_2$O$_3$ samples by optical second harmonic generation [3]. In step two, we corroborated the sensitivity of our experiment to the magnetization induced via the linear magnetoelectric effect. We coated a Cr$_2$O$_3$ sample with a metallic platinum film of 50 nm thickness acting as front electrode and detected the Cr$_2$O$_3$ bulk magnetization induced by 50V applied to the electrodes. This revealed a domain-dependent magnetization one to two orders above our detection limit. In the third step, we repeated the experiment on an uncoated Cr$_2$O$_3$ sample, now employing the charged tip as the source of charge to generate a monopolar magnetic field as described above. We found, however, that the residual Cr$_2$O$_3$ surface roughness of about 4 nm led to a pronounced electrostatic inhomogeneity in this insulating sample that obscured any response expected from the magnetic-monopole field. No signal difference was detected at the position of the antiferromagnetic domain boundaries.

**B. Scanning SQUID magnetometry**

Another possible technique for measuring the induced monopolar field could be scanning SQUID magnetometry. When a charge $n_e \times |q|$ is placed on the magnetoelectric surface, we have seen that the resulting monopole is given by

$$m \approx -\frac{\mu_0}{4\pi \epsilon_0 (\mu + \mu_0)} \frac{q (\alpha_\perp + \alpha_\parallel)}{(\alpha_\perp + \alpha_\parallel)^2}$$

$$= n_e \cdot 1.92 \cdot 10^{-22}\text{Tm}$$

for the case of Cr$_2$O$_3$. The magnetic flux from the magnetic monopole through a Josephson junction is then given by (see derivation in Appendix)

$$\Phi = \int B dz = m(z + d) \int_0^{2\pi} d\Phi \int_0^R r dr \left( \frac{1}{r^2 + (z + d)^2} \right)^{3/2}$$

$$= 2\pi m \left( 1 - \frac{z + d}{\sqrt{R^2 + (z + d)^2}} \right)$$

where $m$ is the magnetic monopole moment, $z$ is the distance of the pickup from the interface, $d$ is the distance of the charge from the interface, and $R$ is the radius of the loop. One of the key challenges in this experiment would be to find a way to fix and localize charge above the surface.

**V. CONCLUSIONS**

In summary, we derived the form of the electric and magnetic fields that are induced by an electric charge above a surface of a semi-infinite slab of magnetoelectric material. We found that, for both isotropic and uniaxial magnetoelectrics, the electric charge induces a magnetic image charge, which is the source of a monopolar field decaying with $r^2$ in the vacuum region. In addition, we showed that the strength of the induced field depends on the value of the isotropic part $\frac{1}{2} (\alpha_\perp + \alpha_\parallel)$ of the magnetoelectric tensor and that a field generated by any anisotropic component of the magnetoelectric response vanishes at the interface. We showed that the magnitude of the response induced by a single electronic charge is large enough to be detectable experimentally, and described searches using muon spin spectroscopy and magnetic force microscopy. Our muon spin spectroscopy data, while not fully conclusive, are consistent with the existence of the monopolar field. We hope that our encouraging initial results, as well as our discussion of other possible experimental approaches for measurement of the monopole, motivate further studies.

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While only 11 of the 58 magnetic point groups that allow the magnetoelectric effect have uniaxial symmetry, in many of the other cases, the tensor can be transformed into a form with a diagonal component and our analysis remains relevant for an appropriate choice of surface cut.
Appendix A: Detailed solution for the isotropic case

From the electrostatic boundary conditions and equations (II B.1, II B.2) it follows that \( m'' = m' \). Here we use cgs units for simplicity. and that \( q'' = q + q' \)

\[
m'' = m' \\
q'' = q + q'
\]

From the second and third boundary condition it is found that

\[
eq q - q' \\
\mu m' + \alpha q'' = -m
\]

Thus we find the following matrix

\[
\begin{pmatrix}
1 & -1 & 0 \\
\epsilon & \alpha & \mu + 1
\end{pmatrix}
\begin{pmatrix}
q' \\
m'
\end{pmatrix}
= \begin{pmatrix}
-q \\
q
\end{pmatrix}
\]

Using gaussian transformations the following form can be found

\[
\begin{pmatrix}
1 & -1 & 0 \\
\epsilon & \alpha & \mu + 1
\end{pmatrix}
\begin{pmatrix}
q'' \\
m'
\end{pmatrix}
= \begin{pmatrix}
-2q \alpha \\
2q \alpha
\end{pmatrix}
\]

Thus

\[
m' = -\frac{2q \alpha}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

and

\[
q'' = \frac{1}{\epsilon + 1} \left( \frac{2q \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2} + 2q \right) = \frac{2q(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

And further

\[
q' = -q + \frac{2q(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2} = \frac{q(\mu + 1)(\epsilon - 1) - \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

Using the previous results one finds the potentials

\[
\phi_e' = \frac{q}{|r - r_1|} - \frac{q}{|r - r_2|} \frac{(\mu + 1)(\epsilon - 1) - \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
\phi_m'' = \frac{-q}{2\alpha} \frac{(\mu + 1)(\epsilon + 1) - \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
\phi_e'' = \frac{q}{|r - r_1|} \frac{2(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
\phi_m' = \frac{q}{2\alpha} \frac{2(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

Where \( \phi \) are electric \( (e) \) and magnetic \( (m) \) potentials inside \( (i) \) and outside \( (o) \) the magnetoelectric slab. Taking the gradients, this leads to the fields:

\[
E^o(r) = \frac{q(r - r_1)}{|r - r_1|^{3/2}} - \frac{q(r - r_2)}{|r - r_2|^{3/2}} \frac{(\mu + 1)(\epsilon - 1) - \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
H^o(r) = -\frac{\frac{2}{\alpha}}{|r - r_2|^{3/2}} \frac{(\mu + 1)(\epsilon + 1) - \alpha^2}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
E^i(r) = \frac{q(r - r_1)}{|r - r_1|^{3/2}} \frac{2(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

\[
H^i(r) = \frac{q(r - r_1)}{|r - r_1|^{3/2}} \frac{2(\mu + 1)}{(\mu + 1)(\epsilon + 1) - \alpha^2}
\]

Appendix B: Detailed solution for the uniaxial case

To solve the problem of a charge adjacent to a slab of uniaxial material one starts with the coupled equations inside the magnetoelectric in the absence of free charge. (We use cgs again for simplicity).

\[
(\varepsilon \perp \nabla_\perp + \varepsilon \parallel \nabla_\parallel)E + (\alpha \parallel \nabla_\perp + \alpha \parallel \nabla_\parallel)H = 0
\]

\[
(\mu \parallel \nabla_\perp + \mu \parallel \nabla_\parallel)H + (\alpha \parallel \nabla_\perp + \alpha \parallel \nabla_\parallel)E = 0
\]

where \( \nabla_\parallel = \left( \begin{array}{c} 0 \\ \frac{\partial}{\partial z} \end{array} \right) \) and \( \nabla_\perp = \left( \begin{array}{c} \frac{\partial}{\partial x} \\ \frac{\partial}{\partial y} \\ 0 \end{array} \right) \) are parts of the \( \nabla \)-operator which are anti-parallel and parallel to the anisotropy axis.

Taking the partial fourier transform along \( x \) and \( y \), i. e.

\[
F(x, y, z) = \frac{1}{4\pi^2} \int \int dk_x dk_y F(k_x, k_y, z) e^{i k_x x} e^{i k_y y}
\]

\[
F(k_x, k_y, z) = \int \int dx dy F(x, y, z) e^{-ik_x x} e^{-ik_y y}
\]

The Fourier transformed magnetoelectric differential equations in terms of magnetic and electric potentials is then given by

\[
\left( \frac{\mu \parallel}{\alpha \parallel} \frac{\alpha \parallel}{\varepsilon \parallel} \right) \left( \begin{array}{c} \phi_m'' \\ \phi_e'' \end{array} \right) = k^2 \left( \frac{\mu \perp}{\alpha \perp} \frac{\alpha \perp}{\varepsilon \perp} \right) \left( \begin{array}{c} \phi_m \\ \phi_e \end{array} \right)
\]

where \( k^2 = k_x^2 + k_y^2 \) and the \( ' \) indicates the derivative with respect to \( z \). Multiplying with the inverse of the first matrix and diagonalizing the equation we find that

\[
\left( \begin{array}{c} \phi_m'' \\ \phi_e'' \end{array} \right) = \frac{k^2}{\mu \parallel \varepsilon \parallel} \left( \begin{array}{ll} \mu \parallel \varepsilon \parallel - \mu \perp \varepsilon \parallel \alpha \perp - \mu \parallel \alpha \perp & \mu \parallel \alpha \perp - \mu \perp \alpha \parallel \alpha \perp - \mu \parallel \alpha \parallel \\ -\alpha \parallel \mu \perp + \mu \parallel \alpha \perp & -\alpha \parallel \mu \parallel - \mu \perp \alpha \parallel - \mu \parallel \alpha \perp \end{array} \right) \left( \begin{array}{c} \phi_m \\ \phi_e \end{array} \right)
\]
Diagonalizing this equation we obtain the eigenvalues

\[
\begin{align*}
\lambda_1 &= -k \sqrt{-\gamma + a + d} \\
\lambda_2 &= k \sqrt{-\gamma + a + d} \\
\lambda_3 &= -k \sqrt{\gamma + a + d} \\
\lambda_4 &= k \sqrt{\gamma + a + d}
\end{align*}
\]

where we substituted

\[
a = \frac{\mu_1 \varepsilon_\parallel - \alpha_\parallel \alpha_\perp}{\mu_\parallel \varepsilon - \alpha_\perp^2},
\]

\[
b = \frac{\varepsilon_\parallel \alpha_\perp - \alpha_\parallel \varepsilon_\perp}{\mu_\parallel \varepsilon - \alpha_\perp^2},
\]

\[
c = \frac{-\alpha_\parallel \mu_\parallel + \mu_\parallel \alpha_\perp}{\mu_\parallel \varepsilon - \alpha_\perp^2},
\]

\[
d = \frac{\varepsilon_\parallel \mu_\parallel - \alpha_\parallel \alpha_\perp}{\mu_\parallel \varepsilon - \alpha_\perp^2} = \sqrt{a^2 - 2ad + 4bc + d^2}
\]

The Eigenvectors are given by:

\[
v_1 = \begin{pmatrix} 1 \\ \frac{\sqrt{2b} \sqrt{a + d - \gamma}}{k (a - d + \gamma) - 2bc} \\ -1 \frac{\sqrt{2b} \sqrt{a + d + \gamma}}{k (a - d + \gamma) + 2bc} \\ 1 \end{pmatrix}, \quad v_2 = \begin{pmatrix} 1 \\ -\frac{\sqrt{2b} \sqrt{a + d - \gamma}}{k (a - d + \gamma) - 2bc} \\ \frac{2b}{k (a - d + \gamma) - 2bc} \\ 1 \end{pmatrix}, \quad v_3 = \begin{pmatrix} 1 \\ -\frac{\sqrt{2b} \sqrt{a + d + \gamma}}{k (a - d + \gamma) + 2bc} \\ \frac{2b}{k (a - d + \gamma) + 2bc} \\ 1 \end{pmatrix}, \quad v_4 = \begin{pmatrix} 1 \\ \frac{\sqrt{2b} \sqrt{a + d - \gamma}}{k (a - d + \gamma) - 2bc} \\ \frac{2b}{k (a - d + \gamma) - 2bc} \\ 1 \end{pmatrix}
\]

Since the potential should not diverge for \( z \to -\infty \), \( C_1 \) and \( C_4 \) are zero, which means that the solution can be written as a combination of the second and the fourth eigenfunctions \( v_2 \) and \( v_4 \).

\[
\phi_m = -C_2 \frac{1}{k} \sqrt{2b} \sqrt{a + d - \gamma} e^{k \sqrt{a + d + \gamma} z} + C_4 \frac{1}{k} \sqrt{2b} \sqrt{a + d + \gamma} e^{k \sqrt{a + d + \gamma} z}
\]

\[
\phi_e = C_2 \frac{1}{k} \sqrt{2b} \sqrt{a + d - \gamma} e^{k \sqrt{a + d + \gamma} z} + C_4 \frac{1}{k} \sqrt{2b} \sqrt{a + d + \gamma} e^{k \sqrt{a + d + \gamma} z}
\]

In the vacuum half space the Maxwell equations reduce to

\[
\nabla \cdot \mathbf{E} = 4\pi q \delta (\mathbf{r} - \mathbf{r}_0)
\]

\[
\nabla \cdot \mathbf{H} = 0
\]

Fourier transforming in the \( xy \)-plane we obtain

\[
\nabla^2 \phi_m(k_x, k_y, z) - (k_x^2 + k_y^2) \phi_m(k_x, k_y, z) = 4\pi q \delta(z - z_0)
\]

\[
\nabla^2 \phi_e(k_x, k_y, z) - (k_x^2 + k_y^2) \phi_e(k_x, k_y, z) = 0
\]

The general solutions to these equations in fourier space are given by

\[
\phi_{m}^{\text{vac}} = D_1 e^{-k(z+z_0)} + \frac{2\pi q}{k} e^{-k|z-z_0|}
\]

\[
\phi_{e}^{\text{vac}} = D_2 e^{-k(z+z_0)}
\]

Applying the inverse fourier transform we find that

\[
\phi_{m}^{\text{out}}(x, y, z) = \frac{q}{\sqrt{x^2 + y^2 + |z - z_0|^2}} + \frac{D_2}{2\pi \sqrt{x^2 + y^2 + |z + z_0|^2}}
\]

Potential of the point charge

\[
\phi_{m}^{\text{out}}(x, y, z) = \frac{1}{2\pi \sqrt{x^2 + y^2 + |z + z_0|^2}}
\]

Magnetic image charge

outside the material, and inside the material,

\[
\phi_{m}^{\text{in}}(x, y, z) = \frac{C_2}{2\pi} \frac{\sqrt{2} \sqrt{a + d - \gamma}}{d(a - d + \gamma) - 2bc} \sqrt{x^2 + y^2 + |z - z_0|^2}
\]

\[
+ \frac{C_4}{2\pi} \frac{2b}{d(a - d + \gamma) + 2bc} \sqrt{x^2 + y^2 + |z - z_0|^2}
\]

\[
\phi_{e}^{\text{in}}(x, y, z) = \frac{C_2}{2\pi} \frac{\sqrt{2} \sqrt{a + d - \gamma}}{d(a - d + \gamma) - 2bc} \sqrt{x^2 + y^2 + |z - z_0|^2}
\]

\[
+ \frac{C_4}{2\pi} \frac{2b}{d(a - d + \gamma) + 2bc} \sqrt{x^2 + y^2 + |z - z_0|^2}
\]

One now can solve the system of equations for the constants by imposing the electromagnetic boundary conditions.

**Appendix C: Effect of anisotropy on the monopolar field**

From Eqn. (24) we see that the strength of the magnetic monopolar field is determined by the parameter \( c_{61}^{\text{int}} \), which has a functional dependence on the three tensors \( \bar{\varepsilon}, \bar{\mu} \) and \( \bar{\alpha} \). To understand this dependence we next analyze the magnitude of \( c_{61}^{\text{int}} \) as we vary the three response functions individually.

First we investigate the dependence on the anisotropy in the magnetoelastic response \( \alpha_0 \) and \( \alpha_1 \), with \( \varepsilon \) and \( \mu \) set equal to isotropic values. In Fig. 10 (a) we show \( c_{61}^{\text{int}} \) as a function of \( \gamma \), which is the scaling between \( \alpha_0 \) and \( \alpha_1 \), such that \( \alpha_1 = \gamma \alpha_0 \) for fixed \( \alpha_0 \). We see that the monopolar field grows linearly with \( \alpha_1 \) and vanishes for \( \alpha_0 = -\alpha_1 \). The orange line shows the change in monopole by keeping the
sum of the components constant but varying the weight, thus $\alpha_\perp = t\alpha_0$, $\alpha_\parallel = (1 - t)\alpha_0$. Interestingly, here the monopolar field strength remains independent of $t$, indicating that it is determined by the sum of both components rather than their relative magnitudes.

Next we discuss the effect of the permittivity tensor on the field strength (the dependence on the permeability is analogous and we do not show it here), with the magnetoelectric response set to a isotropic value. In Fig. 10 (b)) we plot the change in $c_{b1}^{\text{out}}$ when we linearly increase the perpendicular component $\varepsilon_\perp = t\varepsilon_0$ while keeping $\varepsilon_\parallel$ constant and we show the general trend that the strength of the monopolar field decreases when $\varepsilon$ increases. This is because a higher dielectric screening decreases the electric field inside the magnetoelectric which leads in turn to a reduced image monopole strength. Additionally we consider the relative distribution of both components we where we put $\varepsilon_\perp = t\varepsilon_0$, which shows that the higher the total dielectric tensor, the smaller the field. From $\varepsilon_\parallel = (20 - t)\varepsilon_0$, we find that the monopolar field reaches a minimum for a isotropic tensor $\varepsilon$, while a higher anisotropy increases the monopolar field. We find that the strength of the field increases symmetrically independent which component of $\varepsilon$ is increased and decreased if we keep the sum constant.

We know consider the situation in which we have anisotropy in $\alpha$ (Fig. 10 (c)). In this case the symmetry is broken and we find that the anisotropy in $\varepsilon$ now inversely influences how much each element of the magnetoelectric tensor contributes how much to the strength of the monopolar field. Thus, increasing $\varepsilon_\parallel$ will lead to a reduced contribution of $\alpha_\parallel$, which we can see in the plot. In fact, we see that if we increase one element of $\varepsilon$ enough compared with the other, we can even change the sign of the monopole (if the magnetoelectric tensor has elements with different signs) as can bee seen in Fig. 10(c) at $t = 17$. 

\[ a) \alpha_\perp = t\alpha_0, \alpha_\parallel = (1 - t)\alpha_0 \]

\[ b) \varepsilon_\perp = t\varepsilon_0, \varepsilon_\parallel = (20 - t)\varepsilon_0 \]

\[ c) \varepsilon_\perp = t\varepsilon_0, \varepsilon_\parallel = 10\varepsilon_0 \]
Appendix D: LE-\(\mu\)-SR spectra

Here we present representative raw data of our muon spectroscopy measurements presented in section III B for the example point with a stopping energy of 10 keV with an applied field of +10 mT.

![Graph of LE-\(\mu\)-SR spectra](image)

FIG. 11. Typical LE-\(\mu\)SR spectra (20 K, 10 keV, +10 mT) obtained for four positron detectors arranged around the sample. The solid lines are fits to the raw data with an exponential envelop function, i.e. assuming a Lorentzian field distribution.

![Graph of FFT of \(\mu\)SR data](image)

FIG. 12. Averaged FFT power of the \(\mu\)SR raw data (points) and of fits (solid line) shown in Fig. 11.

Appendix E: Magnetic flux through a SQUID loop

Taking the usual form for the magnetic flux through a loop,

\[ \Phi = \int_S \mathbf{B} \cdot d\mathbf{S} \quad \text{(E1)} \]

and the magnetic field that we derived for a monopole at position \(x = 0, y = 0, z = -d\),

\[ \mathbf{B} = \frac{m}{(x^2 + y^2 + (z + d)^2)^{3/2}} \quad \text{(E2)} \]

we integrate along the surface parametrized by

\[ \{ (x, y) \in S | x^2 + y^2 \leq R^2, z = z \} \]

and obtain

\[ \Phi = m \int_S dS \frac{(z + d)}{(x^2 + y^2 + (z + d)^2)^{3/2}} \quad \text{(E3)} \]

\[ = m(z + d) \int_0^{2\pi} \int_0^R sd\phi \left( \frac{1}{r^2 + (z + d)^2} \right)^{3/2} \cdot \text{(E4)} \]

Substituting \( s = r^2 + (z + d)^2 \) and using \( dr = \frac{ds}{2r} \) leads to

\[ \Phi = 2\pi m \frac{(z + d)}{2} \frac{R^2 + (z + d)^2}{(z + d)^{3/2}} \cdot \text{(E5)} \]

\[ = 2\pi m \left( 1 - \frac{z + d}{\sqrt{R^2 + (z + d)^2}} \right) \cdot \text{(E6)} \]

Note that in the limit of a large loop radius, \( R \), we find:

\[ \Phi_{R \to \infty} = \frac{4\pi m}{2} \quad \text{(E7)} \]

which is half the flux created by the point charge as expected from Gauss’ Law.