ac Wien Effect in Spin Ice, Manifest in Nonlinear, Nonequilibrium Susceptibility

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The Wien effect is a model process for field-induced charge creation. Here it is derived for a nonelectrical system: the spin ice "magnetolyte"—a unique system showing perfect charge symmetry. An entropic reaction field, analogous to the Jaccard field in ice, opposes direct current, but a frequency window exists in which the Wien effect for magnetolyte and electrolyte are indistinguishable. The universal enhancement of monopole density speeds up the magnetization dynamics, which manifests in the nonlinear, nonequilibrium ac susceptibility. This is a rare instance where such effects may be calculated, providing new insights for electrolytes. Experimental predictions are made for $Dy_2Ti_2O_7$ spin ice.

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Introduction.—The nonlinear response to an applied field can be as varied as it is interesting—even for the vacuum of quantum electrodynamics, a sufficiently strong electric field will produce electron-positron pairs via the Schwinger mechanism [1,2]. More complex collective "vacua" are provided by weak electrolytes, whose conductivity enhancement via the Wien effect also involves pair creation and unbinding, as first analyzed by Onsager [3].

Our work addresses the Wien effect as a phenomenon occurring in the class of magnetic materials known as spin ice [4], as well as studying in detail its ac version, also applicable to conventional electrolytes. In spin ice, the emergent (and magnetic) Coulomb charges [5] are attached to "Dirac strings" carrying fluxes of the corresponding emergent gauge fields [6]. We find that this completely destroys the steady state known from electrolytes but leaves behind a frequency window in which an analogous quasisteady state can be observed. We provide a combined quantitative analytical-numerical study of the time-dependent Wien effect. Our study extends the conventional theory to include the effect of vacuum polarization, such as occurs, for example, in water ice, via the Jaccard field [7,8], as well as exposing the dynamical suppression of the Wien effect at high frequency.

The charge creation process of the Wien effect couples to a nonlinear susceptibility, providing a clear signature of the field-driven changes to internal correlations. Our search for its theoretical description serves as a case study, as nonlinear susceptibilities, whether related to higher harmonics or to high-field response, are indispensable macroscopic signatures of evolution beyond linear response as seen in optics [9], glassy systems [10–13], liquids [14], superconductors [15], heavy fermions [16], and magnets [17,18].

We thus obtain a description of the nonlinear susceptibility of a spin ice over an unprecedented window of frequencies, amplitudes, and temperatures—a rare possibility for any nontrivial magnet. This can be probed naturally via measurement of the uniform ac susceptibility. The wide parameter range of our predictions also underscores the role of spin ice as an electrochemical model system, providing in particular a perfectly symmetric electrolyte.

The underlying mechanism of the Wien effect is the generation by an external field of an excess—often very sizable—of free charge, as shown in Figs. 1(a) and 1(b) for both a lattice electrolyte and its magnetic equivalent, the magnetolyte. The excess magnetic charge is seen, in turn, to amplify the magnetic response. In the following, we introduce the model magnetolyte, demonstrating that it exhibits quantitatively these key features. We then develop a kinetic model which accurately reproduces both the Wien effect and the magnetic response observed in simulations. Finally, we propose an experimental protocol for the nonlinear susceptibility and discuss the optimal experimental setting for detecting the ac Wien effect in $Dy_2Ti_2O_7$.

Model.—Spin ice consists of a network of corner-sharing tetrahedra of Ising-like magnetic moments (spins) constrained to point along the axis connecting the centers of neighboring tetrahedra, which in turn define a diamond lattice of constant a. Exchange and dipolar interactions are of similar magnitude [19]. However, for configurations satisfying the ice rules of two spins pointing in and two out of each tetrahedron, dipolar interactions maintain an approximate degeneracy. This becomes exact in the dumbbell model [5], in which spins are replaced by magnetic charge dumbbells that touch at the centers of the tetrahedra. Monopoles on top of this vacuum represent a violation of the ice rules with three spins in and one out, or

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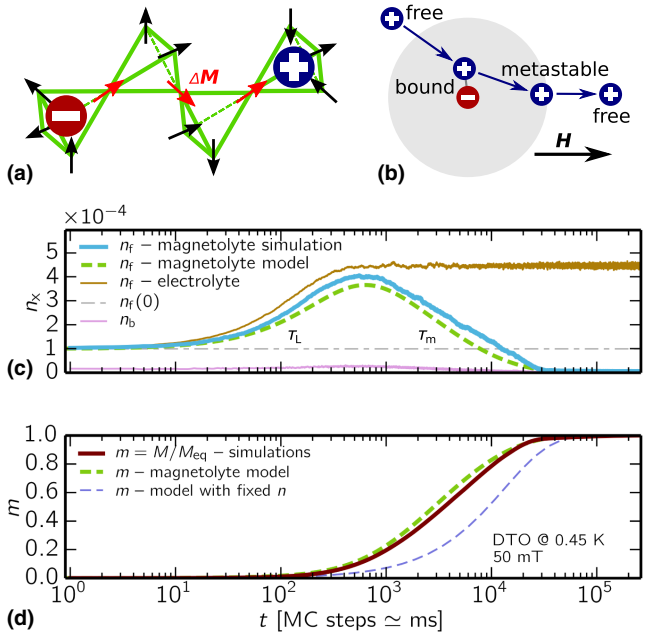

FIG. 1 (color online). (a) Monopoles move via spin flips, and their current magnetizes the ice manifold. (b) The second Wien effect involves the field enhanced dissociation of bound pairs. Nonlinear response: (c) After a field quench, the Wien effect increases the free charge density n_f in an electrolyte. (d) In a magnetolyte with the same initial density and temperature, the free monopole density increase is only transient, counteracted by the growing magnetization m of the system. The increased monopole density is observable in the faster rate of magnetization m compared to a magnetization process at fixed density n. The response is well described by our kinetic model. The bound charge density is only weakly influenced (n_b). Magnetolyte parameters are $T = 0.45$ K, $n_{\mathrm{tot}}(0) \simeq 1.1 \times 10^{-4}$, $n_f(0) \simeq 1.0 \times 10^{-4}$, and $\mu_0 H_0 = 50$ mT; electrolyte parameters are set to obtain the same zero-field density.

three-out-one-in, corresponding to magnetic charges $Q_m = \pm 2\mu/a$ [Fig. 1(a)]. Doubly charged monopoles (four-in or four-out tetrahedra) are costly in energy and can be neglected over the temperature range considered here [20].

The dumbbell model, which we use for both analytics and numerics, leads to a great simplification of the treatment of the dense network of magnetic moments. The energetics are accounted for by the magnetic Coulomb interaction $U(r) = \pm\mu_0 Q_m^2/4\pi r$ [5] between monopoles, arising from dipolar coupling of spins, along with their chemical potential ν [20] (from exchange and dipolar coupling). The emergence of the monopoles from the string background imposes constraints on their motion, leading, e.g., to a renormalization of the diffusion constant D [21] by a factor of $\frac{2}{3}$ compared to a lattice electrolyte [22]. It also accounts for the entropics of spin ice [8,23] as the monopole motion changes the local magnetization. The dumbbell model thus formulates spin ice as a true Coulomb liquid of magnetic monopoles—a magnetolyte—which has the added richness of configurational entropy in the string background. It has been shown to describe the equilibrium properties of spin ice materials such as $Dy_2Ti_2O_7$ (DTO) and $Ho_2Ti_2O_7$ [21,24–26].

We simulate the dumbbell model with periodic boundary conditions and Ewald-summed Coulomb interactions [27]. After equilibration in zero field from the initial ordered configuration with both single-spin flip and worm Monte Carlo algorithms [28–30], the sample evolves via local moves following a chosen field protocol. The locality of the moves ensures physical diffusive dynamics [26,31–33]. The parameters used in our simulations are extracted from experiments on DTO [26].

Wien effect and nonlinear response.—In weak electrolytes, applying an external electric field increases the density of mobile ions following the enhanced dissociation of bound pairs—the second Wien effect. By analogy one would expect the Wien effect to occur in a weak magnetolyte [4] such as DTO below ~1.5 K, where $\mu_0 Q_m^2/4\pi a > 2k_B T$ [34,35]. Our first and central result is that our simulations do indeed show the Wien effect in the increase in monopole density [Fig. 1(c)]. However, this increase is only transient because the monopole currents magnetize the system [Fig. 1(d)], which eventually halts the Wien effect and even reduces monopole density for unrelated energetic reasons (see the Supplemental Material [36]). Crucially, for the temperatures of interest where n_f is small, the magnetization relaxation time τ_m is longer than the Langevin time lag [3] τ_L, over which the Wien effect drives the density increase. Hence, periodic switching of the field direction stabilizes the density increase [Fig. 2(a)] when the switching period falls between these two scales, and even for higher frequencies as we discuss in Fig. 3(c) below. The amplitude dependence is staggeringly similar to that for the electrolyte in constant field [Fig. 2(c)].

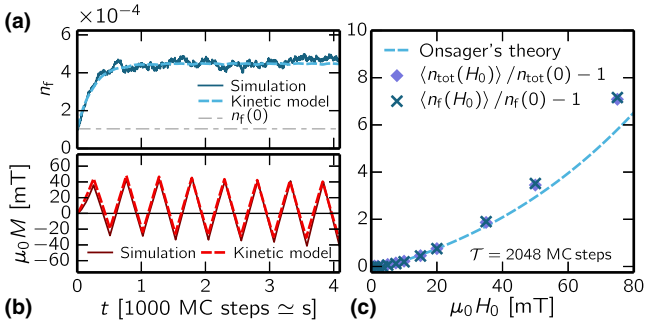

FIG. 2 (color online). (a) Square wave driving stabilizes the free monopole density increase due to the second Wien effect (b) if the magnetization M stays well below $M_{\mathrm{eq}} = \chi_T H_0$. The kinetic model captures the response including the transition from equilibrium to a periodic steady state. (c) The amplitude dependence of the average density increase matches the dc Wien effect theory with no free parameters, confirming spin ice's dynamical window of electrolyte behavior. Magnetolyte parameters are $T = 0.45$ K, $n_{\mathrm{tot}}(0) \simeq 1.1 \times 10^{-4}$, $n_f(0) \simeq 1.0 \times 10^{-4}$, and $\mu_0 H_0 = 50$ mT (a),(b).

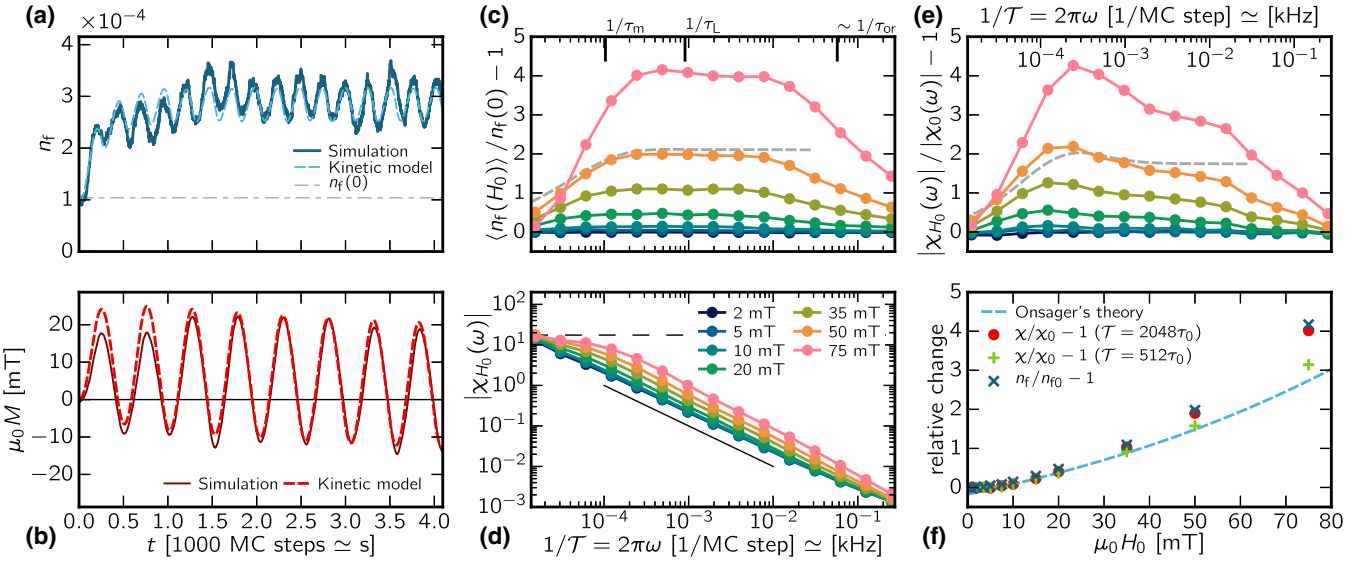

FIG. 3 (color online). (a) The free monopole density increase due to sine driving (b) enhances the magnetic response. (c) The Wien effect persists over a range of frequencies. (d) The enhanced density leads to an increase in the absolute value of the nonlinear susceptibility; the dashed black line is χ_T and full black line is $\propto 1/\omega$. The relative change in χ_{H_0} is shown in (e) revealing additional features in the Wien effect plateau compared to the density increase. (f) The amplitude dependence stays close to Onsager's theory of the dc Wien effect (with mean modulus of the field $\langle|H_0 \sin(\omega t)|\rangle = 2H_0/\pi$) despite the approximations made. The kinetic model (results for $\mu_0 H_0 = 50$ mT) captures the time evolution of density and magnetization [dashed lines in (a),(b)], the low-frequency ransition in density and susceptibility [grey dashed lines in (c) and (e)], and the structure of the susceptibility increase. However, it does not include the high-frequency cutoff due to pair reorientation. Magnetolyte parameters are $T = 0.45$ K, $n_{\rm tot}(0) \simeq 1.1 \times 10^{-4}$, and $n_f(0) \simeq 1.0 \times 10^{-4}$.

Chemical kinetics of monopoles.—The backbone of our dynamical analysis is a pair of coupled rate equations, for the monopole density (4) and for the magnetization (7) whose derivation we sketch in turn.

As in the case of weak electrolytes, monopoles in spin ice at low temperatures can be separated into free monopoles and bound (Bjerrum) pairs, treated as distinct chemical species [34]. Combining this with the creation of bound pairs from the ice manifold (quasiparticle vacuum), we have a double equilibrium: vacuum$\rightleftharpoons_K$ bound$(n_b)\rightleftharpoons$free$(n_f = n_+ + n_-)$ [3,45]. The dissociation constant $K = 2\gamma^2 n_+ n_-/n_b$ controls the equilibrium between the bound and free charges. The activity coefficient γ, found, e.g., from the Debye-Hückel-Bjerrum theory [34,46], gives the modification of the free charge density due to correlations [47].

The external field strongly shifts this equilibrium towards the creation of additional free monopoles. The field reorients and dissociates the bound pairs while the bound charge density is swiftly replenished from the vacuum. Onsager [3] presented an exact solution for the increase in dissociation rate $k_D(b) = F(b)k_D(0)$ and dissociation constant $K(b) = k_D(b)/k_A = F(b)K(0)$, where $b = \mu_0^2 Q_m^3 H_0/8\pi(k_B T)^2$ is linear in a constant applied field H_0, $F(b) = I_1(2\sqrt{2b})/\sqrt{2b} \simeq 1 + b + O(b^2)$, with I_1 the modified Bessel function. The association rate k_A stays constant [3,48].

In dilute electrolytes ($n_f \ll 1$), the steady-state free charge density increases as

$$\Delta n_f(b)/n_f(0) = [\gamma(0)/\gamma(b)]\sqrt{F(b)} - 1. \qquad (1)$$

At fields sufficiently strong to remove the screening atmosphere [49,50], $\gamma(b) \to 1$ ("Onsager's theory"), valid above a field of ~3 mT in Figs. 2(c) and 3(f); in lower fields, a crossover in $\gamma(b)$ from unity to the zero-field value occurs; see also Ref. [51] and the Supplemental Material [36].

Magnetic monopoles in spin ice react to a force $F_m = \mu_0 Q_m(H - M/\chi_T)$ [8], with H the internal field along the [001] axis and $\chi_T = \sqrt{3}\mu_0 Q_m^2/8k_B T a$ the isothermal susceptibility [8,52]. The term M/χ_T expresses the entropic bias towards states with low magnetization. In the following we define $H(t) - M/\chi_T = H_0[h(t) - m]$ with $h(t) = H(t)/H_0$ and $m = M/M_{\rm eq} = M/(\chi_T H_0)$ and work in the limit of fast equilibration of bound pairs with the vacuum: $n_b(H) = n_b(0)$. As discussed in Refs. [3,22,51] the kinetics of the Wien effect in this limit are

$$dn_f/dt = F[b|h(t) - m|]k_D^*(0)(1 - n_f) - k_A n_f^2/2, \qquad (2)$$

where $k_D^* = k_D k_{\rm cr}/(k_{\rm cr} + k_{\rm an})$ represents the effective single equilibrium dissociation rate, which is rescaled by monopole creation ($k_{\rm cr}$) and annihilation ($k_{\rm an}$) rates of the

vacuum-pair equilibrium [51]. Equation (2) may be linearized in b and $n_f = n_f^0 + \Delta n_f$ to give

$$d\Delta n_f/dt = k_D^*(0) b |h(t) - m| - k_A n_f^0 \Delta n_f. \quad (3)$$

In terms of $\zeta(t) = \Delta n_f(t)/(b n_f^0/2)$, we obtain our first constitutive equation

$$d\zeta/dt = [|h(t) - m| - \zeta]/\tau_L^{(0)}, \quad (4)$$

with $\tau_L^{(0)} = \tau_0/[\chi_T n_f(0)]$ the linearized Langevin time and where we used $k_A = \chi_T/\tau_0$ [3,53].

Magnetization dynamics.—Changes in magnetization are coupled to the current density of free (mobile) monopoles $j = \partial M/\partial t = Q_m n_f v/\tilde{V}$, where $\tilde{V}$ is the volume per site and $v = \kappa_m F_m = (Q_m D/k_B T) F_m$ the drift velocity. Thus,

$$dm/dt = [h(t) - m]/\tau_m, \quad (5)$$

with $\tau_m = 9 k_B T \chi_T \tilde{V} \tau_0/n_f a^2 \mu_0 Q_m^2 = (3/2)\tau_0/n_f$ and where, as advertised, $\tau_m^{(0)}/\tau_L^{(0)} = 3\chi_T/2 \gg 1$ at low temperatures (in DTO, $\chi_T \simeq 17.5$ at $T = 0.45$ K). Equation (5) implies a susceptibility [8,54]

$$\chi(\omega) = \chi_T/(1 - i\omega\tau_m) = \chi_T/[1 - (3/2) i\omega\tau_0/n_f], \quad (6)$$

which appears Debye-like, but has a relaxation time dependent on monopole density. As $n_f(t)$ is itself time dependent through Eq. (4), the magnetization relaxation rate, at low field, is enhanced by a factor $1 + b\zeta/2$

$$dm/dt = (1 + b\zeta/2)[h(t) - m]/\tau_m^{(0)}. \quad (7)$$

Equations (4) and (7) form our kinetic model: the nonlinear behavior comes dominantly from the absolute value in Eq. (4) and the charge-magnetization coupling term $b\zeta m/2$ in Eq. (7). For quantitative comparisons, we replace $b/2$ with the full expression on the right-hand side of Eq. (1) and $\tau_L^{(0)}$ with $\tau_L = 2\tau_L^{(0)} n_f(0)/n_f(b)$, which restores the field dependence of τ_L predicted in Ref. [3]. The model does not generally permit a solution in closed form, but it is readily integrated numerically and gives quantitative agreement with our numerical data, as shown in Figs. 1, 2(a), 2(b), 3(a), and 3(b).

Nonlinear susceptibility.—In spin ice the magnetic response is more readily observable than changes in monopole density. In Fig. 3 we show that, as for an electrolyte [45,53,55,56], enhanced density is stabilized by harmonic driving $H(t) = H_0 \sin(\omega t)$, permitting observation in ac susceptibility experiments, extended beyond linear response by increasing the amplitude H_0.

This nonlinear susceptibility $\chi_{H_0}(\omega) = M(\omega)/H_0$ compares the magnetic response $M(\omega)$ to the amplitude H_0 of harmonic driving at the same frequency; $M(\omega)$ is obtained by spectral analysis of $M(t)$ in its periodic steady state (details in the Supplemental Material [36]). In the low-field limit, the usual linear susceptibility is recovered. Similarly, the response $M(l\omega)$ at multiples of the base frequency yields higher harmonics $\chi_{H_0}^{(l)}(\omega)$. Both the magnitude of the field response and the occurrence of higher harmonics are characteristic of the Wien effect, as it couples a scalar (density) to the *modulus* of an applied vector field [53]. Specifically, only odd higher susceptibilities are visible due to the occurrence of even harmonics in density.

Harmonic driving stabilizes a plateau of density increase in a frequency window between $\omega_{\rm low} \simeq 1/\tau_m$ and $\omega_{\rm high} \gg 1/\tau_L$ [Fig. 3(c)]. The process limiting the ac Wien effect at high ω is thus the establishment of Wien effect correlations, i.e., the reorientation of bound pairs [57] along the field direction (on a time scale $\tau_{\rm or}$), and not the density relaxation (τ_L).

The Wien effect increase in density translates into an increase in susceptibility as observed in Figs. 3(c)–3(e). In the simplest approximation, the susceptibility follows the density as in Ryzhkin's original noninteracting theory [8], i.e., $\Delta\chi_{H_0}(\omega)/\chi_0(\omega) \overset{\omega \gg \tau_m}{=} \Delta\langle n_f(H_0)\rangle_T/n_f(0)$. We compute $\chi_{H_0}(\omega)$ in our simulations and observe that this approach is remarkably successful [Fig. 3(f)], especially so at frequencies $1/\tau_m \lesssim \omega \lesssim 1/\tau_L$ [Fig. 3(e)] where density fully relaxes as the field changes [even from zero field, as in Fig. 1(c)]. Further, a reduction in magnetic response is observed between $1/\tau_L$ and $1/\tau_{\rm or}$ reflecting the increasing fraction of time the monopoles spend establishing the Wien effect rather than magnetizing the system. The Wien effect effectively vanishes for $\omega \gg 1/\tau_{\rm or}$.

As there are open questions about the exact nature even of the linear response in DTO at $T < T_f \simeq 0.6$ K, at which dynamics slows beyond laboratory time scales [58–62], it is convenient that our approach relies only on measuring relative quantities, eliminating many nonuniversal contributions. As an example, τ_m can be fixed from the linear susceptibility $\chi_0(\omega)$. Moreover, our theory holds above T_f; the 0.7 K equivalent of Figs. 3(e) and 3(f) is given in the Supplemental Material [36]. Finally, we note that our model should contribute to the resolution of open issues concerning the experiment of Ref. [4] (Refs. [63–67]; see Ref. [68] for a summary).

Conclusions.—There exists a time and frequency window where the Wien effect in spin ice is just the same as in a weak electrolyte. Our consequent prediction for the complex magnetic response of spin ice seems hard to imagine without the mapping of a microscopic spin Hamiltonian to the monopole Coulomb gas. Our main proposal for experiment concerns the strong amplitude dependence of a nonlinear susceptibility which serves as a novel observable for the Wien effect.

The results are also the most detailed modeling of the ac Wien effect in any material system. They enable its study in an "electrolyte" which is perfectly symmetric under the interchange of the sign of the charges, and provide access to

more delicate aspects of the second Wien effect such as the reorientational dynamics of bound pairs at high frequencies.

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