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Intrinsic and extrinsic nonstationary field-driven processes in the spin-ice compound $Dy_2Ti_2O_7$

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Nonequilibrium processes are probed by ultrasound waves in the spin-ice material $Dy_2Ti_2O_7$ at low temperatures. The sound velocity and the sound attenuation exhibit a number of anomalies versus applied magnetic field for temperatures below the "freezing" temperature of ∼500 mK. These robust anomalies can be seen for longitudinal and transverse acoustic modes for different field directions. The anomalies show a broad hysteresis. Most notable are peaks in the sound velocity, which exhibit two distinct regimes: an intrinsic (extrinsic) one in which the data collapse for different sweep rates when plotted as function of field strength (time). We discuss our observations in context of the emergent quasiparticles which govern the low-temperature dynamics of the spin ice.

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Systems with competing interactions are of a great interest in many-body physics. For example, in magnetic systems, frustration manifests in a number of interesting properties, distinct from the ones of ordinary magnets or spin glasses.[1] Due to their (thermodynamically) large ground-state degeneracy, frustrated magnets often remain in an apparently disordered state down to the lowest temperatures.

Among the best examples of frustrated magnetic systems are those with a pyrochlore magnetic lattice, which consists of corner-shared tetrahedra with a magnetic ion at each tetrahedron vertex. $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$ belong to the family of rare-earth titanates with the pyrochlore lattice, which have attracted much interest in recent years because of their unusual spin-ice ground state (the arrangement of magnetic moments there is analogous to the proton configuration in water ice), deconfined fractionalized excitations with magnetic Coulomb interactions ("magnetic monopoles"), and dynamics.[2–18] A broad range of experimental techniques have been brought to bear (e.g., magnetic, thermodynamic, and neutron-scattering measurements).[2–20] The application of a uniform magnetic field is particularly rich (as it can act as an effectively staggered field on account of the noncollinear easy axes[21]) and has produced a symmetry-sustaining Kasteleyn transition,[18] dimensional reduction to kagome ice,[7,8,13,14] and a Zeeman equivalent of deflagration.[17]

The aim of our Rapid Communication is to study the low-temperature ($T < 1.5$ K) behavior of $Dy_2Ti_2O_7$ to understand the role of spin-ice physics and monopoles in the low-energy field-induced nonstationary processes in spin ices. We employ ultrasound measurements as these are well known for their outstanding sensitivity to probe various phase transitions and crossover phenomena.[22] In our experiments, the phase-sensitive technique[23] was used to detect the relative change of the sound velocity $\Delta v(T,H)/v$ and the sound attenuation $\Delta\alpha$ as a function of temperature and external magnetic field.

The nonstationary processes observed in field sweeps in spin ice have been argued[17] to arise from an inability of the phonons to carry away the Zeeman energy released by the flipping spins when thermal runaway is triggered due to "supercooling," which arises when the sparseness of defects at low temperatures induces an exponentially slowdown of the dynamics.[9,10,17]

Ultrasound, as a direct probe of the phononic degrees of freedom, thus provides a natural and direct handle on this phenomenon. We find that the sound velocity exhibits sets of well-defined spikes as the field is swept. We analyze the shape of these peaks, which are highly asymmetric on account of the fundamentally distinct nonequilibrium mechanisms involved: an "intrinsic" rise followed by an "extrinsic" fall, evidenced by data collapse for various combinations of peak positions and sweep rates—the former involves the release of (Zeeman) energy from spin, the latter transfer of energy out of the sample. In addition, for the case of an applied field in the [111] direction, we find a very clear signature of the transition to saturated spin ice, including the onset of hysteresis at the monopole "liquid-gas" transition.[24]

Methods. Single crystals of $Dy_2Ti_2O_7$ were grown under oxygen-gas flow by the floating-zone method in an infrared furnace.[25] $Dy_2Ti_2O_7$ has a cubic crystal structure with the space group $Fd\bar{3}m$. The crystallographic orientations were controlled by the x-ray-diffraction Laue technique. The lengths of the samples used in the experiments are 2.57 mm in [111], 3.41 mm in [001], and 1.09 mm in [112] direction, respectively. Resonance $LiNbO_3$ or wide-band PVDF-film (polyvinylidene fluoride film) piezoelectric transducers glued on the polished surfaces of the sample are implemented to generate and detect ultrasound waves in the frequency range of 60–110 MHz. Multiple ultrasonic echoes were observed in the experiment due to ultrasound-wave reflections from the two parallel polished sample surfaces. The sample was fixed in a sample holder made out of brass in a ^{3}He cryostat. A RuO_2 thermometer was attached directly to the sample. The sample was in vacuum and the sample holder was thermally connected to the ^{3}He chamber. We have studied the c_{11} ($\mathbf{k}\|\mathbf{u}\|[001]$), $c_L = (c_{11} + 2c_{12} + 4c_{44})/3$ ($\mathbf{k}\|\mathbf{u}\|[111]$), and $c_T = (c_{11} + c_{44} - c_{12})/3$ ($\mathbf{k}\|[111],\mathbf{u}\perp\mathbf{k}$) acoustic modes. Here $\mathbf{k}$, $\mathbf{u}$ are the wave vector and polarization of the acoustic wave, respectively, and c_{ij} are the elastic moduli. The sound

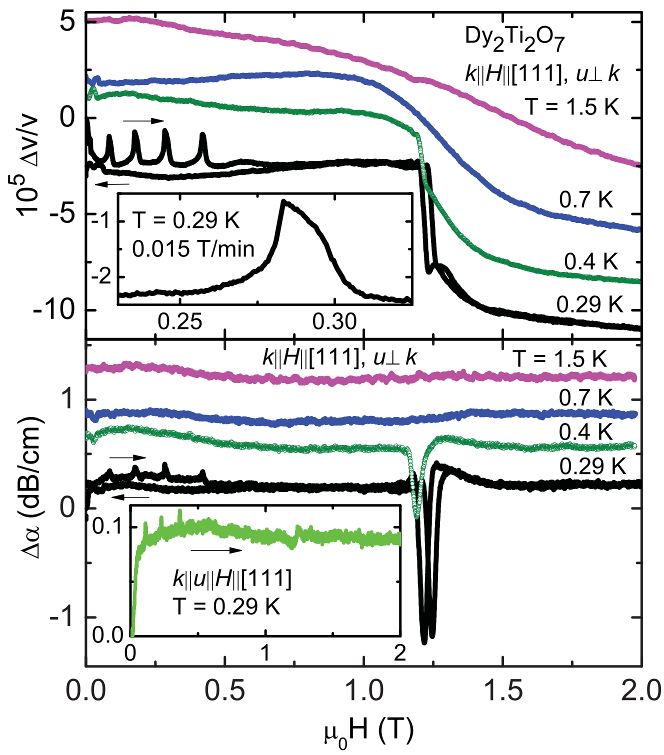

FIG. 1. (Color online) Field dependence of the sound velocity $\Delta v/v$ (top) and the sound attenuation $\Delta\alpha$ (bottom) of the transverse ultrasonic wave propagating along the [111] ($k\|H\|[111]$ and $u\perp k$) direction (acoustic c_T mode) at different temperatures measured under a ZFC condition. Results for up and down field sweeps (indicated by arrows) are shown for the lowest temperature of 0.29 K. All data curves are arbitrarily shifted along the y axis for clarity. The upper inset shows some details for a single peak in the sound velocity. The field dependence of the sound attenuation for the longitudinal mode (acoustic c_L mode) at 0.29 K is given for comparison in the lower inset. Only the c_T mode demonstrates a sharp attenuation anomaly at $\sim$1.25 T.

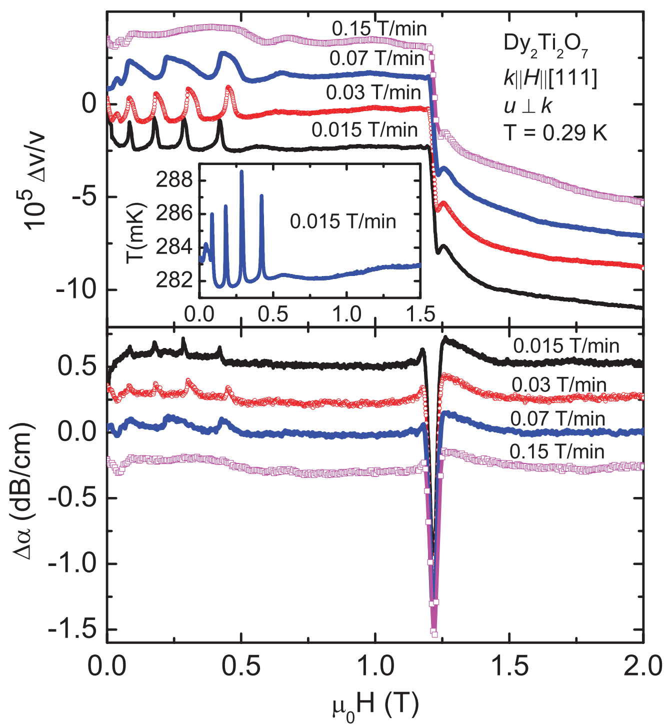

FIG. 2. (Color online) The sound velocity (top) and the sound attenuation (bottom) of the c_T acoustic mode measured at 0.29 K and various field-sweep rates. All data were obtained under a ZFC condition. The data curves are arbitrarily shifted along the y axis for clarity. The inset shows the temperature change, measured during an up field sweep by a RuO_2 thermometer directly attached to the sample.

velocity $v(\mathbf{k},\mathbf{u})$ is related to the elastic modulus via $c = \rho v^2$, where ρ is the mass density of the crystal. We have also performed ultrasound measurements for the [112] direction ($\mathbf{k}\|\mathbf{u}\|[112]$). The applied magnetic field was directed along the sound-propagation direction. Most of the field dependences of the sound velocity and attenuation are reported for the zero-field-cooled (ZFC) conditions.

Results. Figure 1 shows typical field sweeps of the sound velocity and the sound attenuation for the c_T acoustic mode measured at various temperatures. Several peaks appear in both the acoustic characteristics below 0.5 T, in the temperature range of 0.29–0.45 K (c_{11} and c_L acoustic modes show similar features). In addition, there is an abrupt drop for the sound velocity and an anomaly for the attenuation occurs at 1.25 T. It turns out that the attenuation of the longitudinal mode c_L (see the inset in the lower panel of Fig. 1) does not reveal any feature at 1.25 T, while the low-field peaks are present. All features (except for the one at 1.25 T) disappear completely at higher temperatures, $T \geqslant 0.5$ K.

In more detail, we have measured field dependences of the sound velocity using sweep rates in the range of 0.015–0.15 T/min at $T = 0.29$ K (see Fig. 2). This reveals a changeover from narrow peaks to broad peaks by increasing the sweep rate. The maximal height of the peaks for various sweep rates was quite comparable for all peaks. The number of peaks varies little for different field orientations and for different sweep rates (Fig. 2). With increasing sweep rate (and thus peak width), the separation between the peaks decreases, so that peaks merge for large sweep rates; also the positions of the maxima are shifted in field, a clear sign for their nonequilibrium nature. A further indication that we are not probing equilibrium spin configurations is provided by the thermometer installed on the sample, which shows a change of temperature at the values of the field where the peaks of the sound velocity appear[17] (inset of Fig. 2). Due to the nature of the thermometer's contact, the absolute scale of the spikes is likely understated. It appears that the change of sound velocity thus in large part arises from the change in the temperature of the sample at these values of the magnetic field. The phonons thus provide an *in situ* thermometer. However, additional experimental and theoretical work is required in order to convert the sound-velocity change quantitatively into a temperature change.

Before we analyze these findings in more detail, we briefly review the basic model for spin ice.[4] To a good approximation, we are dealing with Ising spins $\sigma = \pm 1$, whose magnetic moment of $|\boldsymbol{\mu}| \approx 10\mu_B$ points along the local easy axis which joins the ion site with the centers of the tetrahedra which share it. All states obeying the ice rules (with two spins pointing into, and two out of, each tetrahedron) are (near-)degenerate ground states.

Tetrahedra violating the ice rules appear as pointlike defects experiencing a mutual relative *magnetic* Coulomb interaction $\frac{\mu_0 Q^2}{4\pi r}$, where $Q \approx 4.6\mu_B/\text{Å}$ is the effective magnetic charge of these monopoles. Single spin flips are only possible at low temperature when they correspond to motion of monopoles—otherwise they are suppressed by an exponentially small Boltzmann factor $\exp[-\Delta/T]$, with $\Delta \approx 4.3$ K. This leads to a strong slowdown of the dynamics below $T_f \approx$ 500–600 mK.[3,9,10,15]

As a result, at low T, field sweeps drive the system out of equilibrium: The magnetization change demanded by the changing field cannot be established sufficiently fast. Instead, "reequilibration" can occur in the form of avalanches with the temperature rising up to T_f,[17] which show up as the peaks in our measurements of the sound velocity.

With this in hand, let us analyze these peaks in more detail. As a starting point, note that when the peaks are well developed (e.g., for the slow sweep rate 0.015 T/min), their overall shape is almost independent of the field at which the peak is triggered (Fig. 3, bottom). Although we have observed the sound-velocity and the sound-attenuation peaks already at 0.015 T/min [the magnetization jumps were detected only above 0.025 T/min (Ref. 17)] stopping a field sweep exactly at the sound-velocity-peak position leads to a relaxation of the sound velocity (and the temperature) to the value characteristic of the valleys between the peaks (not shown), meaning that for much slower sweep rates the peaks should disappear (in agreement with Ref. 17). This can also explain the broadening

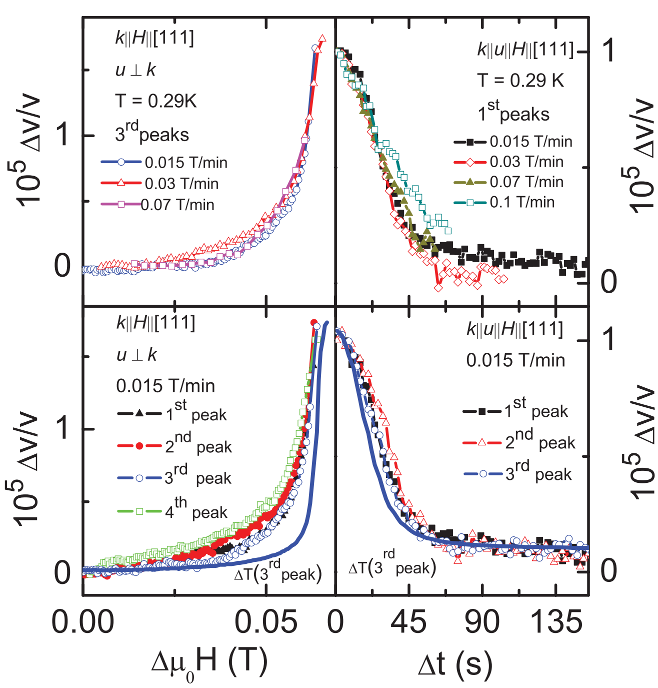

FIG. 3. (Color online) Behavior of the rising parts (left) of the sound-velocity peaks of the c_T acoustic mode for different sweep rates (upper panel) and different peaks (lower panel) as a function of magnetic field at $T = 0.29$ K. The right panels show the descending parts of the velocity peaks for the c_L acoustic mode as a function of time. The blue solid (lower) lines are uncalibrated temperature traces.

of the ultrasound features for the faster field sweeps when the relaxation time approaches the sweep time between the adjacent peaks (see Fig. 2).

Comparing different sweep rates unearths a surprising feature: There appear to exist two qualitatively different regimes. To illustrate this statement we have plotted in the left panels of Fig. 3 the rising parts of the sound velocity peaks with different field-sweep rates (top) and for different peaks (bottom) as a function of the applied field, while in the right panels the descending parts of peaks and the temperature are plotted as a function of time. We have plotted rising parts for the c_T mode, and the descending one for the c_L mode for different sweep rates, for which the collapse is most prominent; the peak shapes of the two modes behave slightly different from each other. Scaling is evident in both panels.

The explanation of the scalings is as follows. The external magnetic field drives the motion of a small number of thermally activated monopoles in $Dy_2Ti_2O_7$ (which leads to a small response rate). For the system's state to keep up with the changing field, more monopoles need to be created so that spins can be flipped at a higher rate. However, energy barriers arise from a competition between the Zeeman energy gain and the cost of creating and separating monopoles, and perhaps also from disorder. Hence, the response has mostly intrinsic character in that it involves a distribution of magnetic energy barriers and not other degrees of freedom such as phonons. The energy released due to the growing number of monopoles moving along the field does not leave the sample quickly, leading to local heating (cf. Fig. 2) resulting in an avalanche:[17] More monopoles get thermally activated, more Zeeman energy is thus released, etc.

On the other hand, the descending parts of the peaks in the sound velocity, i.e., the release of the energy to the outside world, are different in nature: The collapse of the curves as a function of time (regardless of field strength) is consistent with the heat simply disappearing through a thermal contact; indeed, the tail of the peak fits well to an exponential fall-off with a time scale of roughly 1 min. Crucially, the shape of the temperature traced by the thermometer follows this peak decay quite closely, much more so than on the rising side, where the temperature notably lags behind the sound velocity's rise.

Two final remarks on the low-field regime are in order. First, our results crisply complement neutron scattering[12] and the recent magnetization[17] results on nonstationary field sweeps, particularly by providing a detailed and time-resolved focus on the nature of the thermal runaway and subsequent cooldown.

Second, very recent studies[26,27] of the monopole dynamics in $Dy_2Ti_2O_7$ at low temperature suggest that there exist several regimes of monopole relaxation in spin ice, in particular, with slow dynamics below T_f. From Fig. 3, we observe that the relaxation of the sound velocity to the value characteristic of the valleys between peaks is ∼1 min, in the same ballpark but somewhat shorter than a monopole lifetime extracted in Ref. 27, ∼150 s. Notice, however, that relaxation times are observed to be field dependent.[28]

Next, we turn to the strong-field regime, where there is a monopole liquid-gas transition in spin ice. This takes place between the kagome ice regime and the saturated state, which have a low and high density of monopoles, respectively—see

Refs. 5, 8, 13, and 14 for details. For our purposes it is enough to note that the magnetic field applied along the [111] direction acts as a staggered chemical potential for the monopoles.[5] At low temperature, the monopole density exhibits the discontinuous jump of a first-order transition, whereas there is a continuous crossover at higher temperature. A critical endpoint separates the two.[29]

Measured values for the critical field H_c agree with the value $H_c \sim 1.25$ T (see Fig. 1), observed via neutron scattering,[30] however, they disagree with $H_c \sim 0.93$ T, observed in magnetocaloric and magnetization experiments.[24,29] Anomalies are detected in specific-heat measurements at both $\sim$1 and $\sim$1.25 T for $T < 0.3$ K.[31] A careful estimation of the demagnetization effect in our experiments leads to a corrected value of the $H_c = 0.96 \pm 0.04$ T, which is very close to one obtained in Refs. 24 and 29. We also see an onset of the hysteresis, clearly visible at $T = 0.29$ K (Fig. 1) but not at $T = 0.7$ K. In between, at $T = 0.4$ K, $\Delta v/v$ is very steep, suggesting proximity to the critical endpoint.

In passing, we note that our results indicate that the exchange interaction in $Dy_2Ti_2O_7$ is not direct. This is suggested by the fact that the sound attenuation at H_c is stronger in the transverse mode than in the longitudinal one, given that the leading contribution to the exchange-striction coupling is proportional to the scalar product of exchange gradient and the sound polarization.[32]

In conclusion, we have studied nonequilibrium processes and the role of magnetoacoustic interactions in spin-ice $Dy_2Ti_2O_7$ by means of ultrasound measurements. Unusual anomalies have been detected in all acoustic modes studied. The correlation between magnetization plateaus and the peaks in the sound velocity shows that the magnetoacoustic interactions can play an important role in our understanding of the spin ice. We have found that our ultrasound technique is particularly sensitive to these effects, where the periodicity, shape, width, number, and distance between peaks can be crisply extracted. The temperature of the sample follows the sound-velocity change at the peak positions. Field-driven monopoles change the magnetic correlations in the system, and phonons eventually carry the energy out of the system. We thus observe a quasiperiodic change between thermodynamically unstable states through nonequilibrium processes, as in the "bottleneck" effect. It will be interesting to contrast this in detail to the two-dimensional magnetic arrays,[33] where the role of monopoles and avalanche processes in a nonthermal ensemble have been a recent focus of attention.[34]

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