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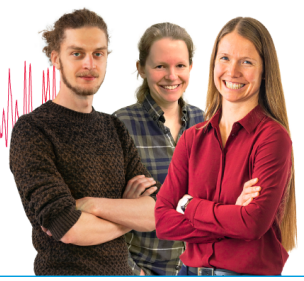

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ABSTRACT

A quasi-binary two-dimensional Ising critical system with the main components D_2O and butyric acid confined by surfactant layers has been studied. The surfactant forms large planar layers and is the basis of the charge density waves with wave fronts aligned with the layers. To orient the domains in an external magnetic field, thulium ions were added to the system (replacing sodium in the surfactant with thulium and adding more $TmCl_3$). The critical behavior of the forward scattering and the correlation length were observed to be more mean-field-like. This can be explained by the presence of the trivalent thulium ions mediating between water and butyric acid. The high-Q scattering could be distinguished in the different directions and the ideal two-dimensional critical composition fluctuation exponent $\eta_{xy} = 1/4$ was observed, while the other exponent $\eta_z = -0.08 \pm 0.06$ was slightly negative due to a finite acceptance angle and the finite magnetic field. The orientationally averaged high-Q exponent x of this study is well-explained by $\eta_{xy} = 1/4$ of the two-dimensional Ising behavior and $\eta_z = 0$.

Recent developments in electrolytes often return to the environmentally friendly and nonflammable water as a base. This is true for lithium batteries[1] and for fuel cells,[2] where water is a by-product. Slightly hydrophobic molecules (such as dimethyl carbonate[3] or TFSI[4] etc.) often come into play as well as polymers that support the formation of the solid electrolyte interphase[4] or the membrane of the fuel cell itself.[2] Apart from that, amphiphilic molecules are used as ion flow stabilizers.[5] Thus, electrolyte formulations include a wide variety of materials for many purposes.

Therefore, the basic understanding of composition fluctuations of water mixed with a slightly hydrophobic molecule as a complex fluid is important, especially when it comes to modifications with surfactant and/or salt. In the literature, such investigations have been carried out experimentally[6–10] and theoretically.[11,12] This kind of fluid consists of water and a slightly hydrophobic substance with a tendency to de-mixing. As a third component, an antagonistic salt or a surfactant is added. The amphiphilic molecules are located at the domain interfaces and give rise to charge density waves (CDW).[13,14] This in turn confines the composition fluctuations to two dimensions with a significantly different Ising criticality (and exponents γ, ν, and η,[15] where γ is connected to the susceptibility, ν to the correlation length, and η to the correlation function) than in three dimensions. In small angle neutron scattering (SANS) studies,[16] the critical correlation function exponent η was suspected to be direction-dependent, i.e., dominated by a confinement in the x and y directions with a different imprint in the third direction.

The classical binary system made of heavy water and butyric acid (BA) has been studied extensively in terms of phase diagrams[17]

and more experiments are carried out with isobutyric acid.[18] The latter also has been studied with respect to critical exponents.[19–21] In one and two dimensions, the Ising model can be solved analytically using the matrix transfer method.[22] Therefore, the critical exponents γ, ν, and η—that we focus on here—are rational numbers,[15] while in three dimensions, they had to be determined numerically. While many experiments confirm the critical exponents in three dimensions,[19–21] the number of experiments is sparse for lower dimensions due to technical obstacles. For magnetic systems,[23,24] there are more varieties, while for composition and density fluctuations the number of experiments is a few only.[6,7,25] With the distance to the critical dimensionality of 4, the critical exponents are growing. Thus, the observables close to the critical point grow much faster in lower dimensions compared to the classical three dimensions. These differences need to be studied in more detail because technical applications need an answer about the details of critical behavior in lower dimensions.

The fundamental understanding of the two-dimensional Ising behavior is highly important for quantum computers[26,27] with the highest efficiencies. Another tightly linked topic is superconductivity[24] for advanced electronic technologies. Therefore, the question of the critical temperature is highly important[28] that lately were also extracted by artificial intelligence algorithms.[29,30] However, the Ising criticality is also connected to societal questions, which evolve from complex networks with interactions.[31] The critical correlation function exponent η has attracted many researchers[32] recently, with questions about the dimensionality.[33] This takes us to the interaction range,[34] which is also linked to the quality of domain interfaces.

As the boundary conditions of the domains are now different along the CDW and perpendicular because of the presence and absence of amphiphile, we expect different corrections by the exponent η in different directions. This anisotropy of the exponent η was the basis for the current study, where we took a critical fluid with surfactant and significantly increased the amount of thulium ions (supplementary material), which allow the domains to be oriented in a magnetic field.[35] We studied our model fluid using small-angle neutron scattering and evaluated the Ising criticality in addition to the high-Q scattering, where the exponent η for the different directions displays.

The basis for the Ornstein–Zernicke-like fluctuations has been described in the work of Fisher.[36] The real space correlation function $G(r)$ is described by the following expression:

$$G(r) = \frac{\exp(-r/\xi)}{r^{d-2+\eta}}. \tag{1}$$

The correlation length ξ describes part of the decay. The other part is given by the power law in the denominator. The exponent is given by integer values of the dimensionality d, the value 2, and the critical correlation function exponent η, which is usually small compared to unity. The scattering function is associated with a Fourier transform. One obtains for three dimensions,

$$S(Q) = \frac{S(0)}{(1+Q^2\xi^2)^{1-\eta/2}}. \tag{2}$$

The integer value of the exponent is related to the exact analytical expression of the Fourier transform. The correction associated with $\eta = 0.04$[21,37] holds in the limit of large scattering vectors Q.

For a quasi-binary liquid with surfactant or antagonistic salt, the critical composition fluctuations are confined to two dimensions between the planar surfactant structures that form the wave fronts of the CDW. Sadakane *et al.*[6] have presented a scattering function derived from the real space correlation function of Onuki.[13,14] The analytical expression is

$$S(Q) = \frac{S(Q=0)}{1+\left\{1-\kappa^2/\left(1+\lambda_D^2Q^2\right)\right\}\cdot\xi^2Q^2}. \tag{3}$$

The Debye length λ_D is derived in the well-known Debye–Hückel theory,[38] which describes the screening of charges in a liquid containing ions. In our case, λ_D has an approximate value of 7 to 9 Å. The parameter κ describes the strength of the CDW correction. It is greater than unity for temperatures far from the critical phase boundary, where the correlation length ξ is small. In our case, the critical fluctuations are so strong that $\kappa < 1$. While the criticality of the forward scattering is still related to the scattering profile at smallest Q, the correlation length ξ is usually obtained at slightly higher Q compared to the classical unconfined three-dimensional Ising system [Eq. (2)]. However, the integer exponent of the power law is still 2.

In a previous publication,[16] we argued that the exponents at large Q are summed of the two-dimensional confined critical behavior and the one-dimensional behavior along the CDW. Such a sum of exponents usually holds only for large aspect ratios of the domains.[39] In our case (as we will see in the following), the domains are only slightly deformed by the surfactant layers, so the net exponent is an average of the dimensions involved. Furthermore, in the previous publication, we assumed a still three-dimensional correlation function. For the two confined dimensions, this is not true and one obtains the asymptotic scattering function,

$$S(Q_{xy}) = \frac{S(0)}{(1+Q_{xy}^2\xi^2)^{3/2-\eta_{xy}}}. \tag{4}$$

For a two-dimensional Ising system, one expects $\eta_{xy} = 1/4$. For the remainder of this manuscript, we call the exponent in the high-Q limit $x = -3 + 2\eta_{xy}$.

The surfactant molecules attract either the water or the butyric acid molecules on either side of the layers. Therefore, the composition profiles at the surfactant layers have a sharp transition from one type of domain to the other. For these structures, the following expression is motivated:

$$S(Q_z) \sim \left|\frac{\sin(Q_z h)}{Q_z h}\right|^{2-\eta_z}, \tag{5}$$

with the domain size $2h$. The corresponding high-Q exponent is $x = -2 + \eta_z$. While an oriented domain structure was desired for our system, achieved by the external magnetic field, in order to separate the influence of the different non-integer exponents, η_{xy} and η_z, an orientational averaging in the x–z plane would result in

$$S(Q) \sim Q^{-5/2+(\eta_{xy}+\eta_z)/2} \tag{6}$$

for the high-Q limit. Therefore, the dominant exponent $-5/2$ is no longer an integer, and the corrections may not be as small compared

to 1/2 as $\eta_{xy} = 1/4$. The corresponding exponent $x = -5/2 + (\eta_{xy} + \eta_z)/2$ holds for the orientational average in the x–z-plane.

While the high-Q limit describes the composition profiles at the interfaces between the domains, the forward scattering $S(0)$ and the correlation length ξ describe the larger scales of the composition fluctuations. Their criticality is related to the Ising model and is described in a universal way,

$$S(0) = S_0 \tau^{-\gamma} \quad \text{and} \quad \xi = \xi_0 \tau^{-\nu}, \tag{7}$$

with the reduced temperature $\tau = |T - T_C|/T_C$ (T_C is the critical temperature), the amplitudes S_0 and ξ_0, and the critical exponents γ and ν. While the first three parameters are material-dependent, the classical values for the exponents $\gamma = 1.239$ and $\nu = 0.629$ are quite universal in three dimensions.[15,40] Usually, still the scaling relation $\gamma/\nu = 2 - \eta$ holds. For curiosity, we mention the hyperscaling $\gamma/\nu = d/2 - \beta$ with β, the critical exponent of the phase separated compositions.[41]

The studied system displays an upper critical solution temperature, as we can see in Fig. 1. For the phase boundaries, we employed viscosity measurements that display critical maximum and SANS measurements that display criticality for the forward scattering and the correlation length. Note that for the whole range, the differences between spinodal and binodal agree within ±0.4 K or better and the statistical noise is larger than that principal difference. For the initial phase diagram, we used 36 wt. % butyric acid vs D_2O and 6.9 mmol/l $TmCl_3$ and 3.7 mmol/l $TmDS_3$. For the final SANS measurements (indicated in red), we increased the $TmDS_3$ concentration to 7.4 mmol/l because the exponents were raised not completely from the 3D to the 2D case ($1.239 < \gamma = 1.51 \pm 0.04 < 1.75$ and $0.629 < \nu = 0.75 \pm 0.02 < 1$). However, the final sample even displayed lower exponents as we will see in the following. The explanation is the controversy trend of growing exponents with more amphiphile but decreasing exponents with more trivalent Tm cations that mediate between the two major liquids.

We will focus our discussion on the anisotropy of the scattering profile at 294 K with a horizontal magnetic field of 2 T.[42] The field direction (z) was perpendicular to the neutron flight path (y)

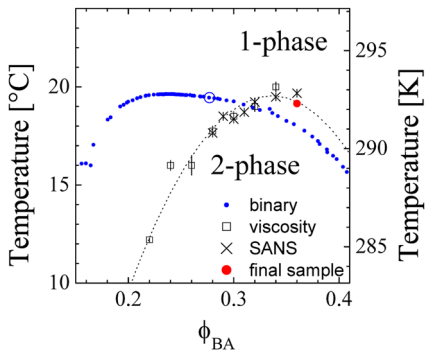

FIG. 1. Experimental phase diagram: temperature vs butyric acid mass fraction. The blue symbols indicate the binary system made of heavy water and butyric acid from Ref. 17. The blue circle indicates the critical point. The system of this manuscript is made of heavy water and butyric acid (at 36 wt. %: 6.9 mmol/l $TmCl_3$ and 3.7 mmol/l $TmDS_3$). The different methods are indicated in the legend. The final sample is indicated by the red dot (here even 7.4 mmol/l $TmDS_3$).

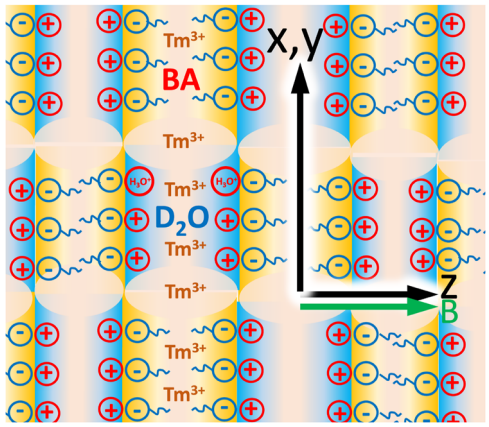

FIG. 2. Illustration of the CDWs in the quasi-binary system of heavy water (D_2O) and butyric acid (BA) with the surfactant thulium dodecyl sulfate ($TmDS_3$) and $TmCl_3$. The domains fluctuate mainly in the x and y planes, parallel to the wave fronts of the CDW and the surfactant layers. The propagation of the CDW is along the z-direction.

and the vertical direction (x) (the discussion will develop the orientation of the domains that we specify in Fig. 2). Sufficient statistics was acquired to display the vertical and horizontal ($\pm 15^\circ$) sections of the scattering profiles (Fig. 3). One observes clear differences at lower $Q < 0.1$ Å^{-1} (with Sadakane fit) and also at highest $Q > 0.1$ Å^{-1} (with a power law fit Q^x). The horizontal correlation length $\xi = 102$ Å is slightly larger compared to the vertical length $\xi = 96$ Å.

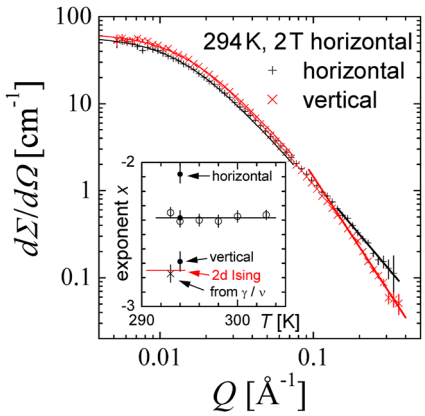

FIG. 3. Horizontal and vertical sections of the full two-dimensional SANS profile with an acceptance of ±15° at an applied horizontal magnetic field of $B = 2$ T and a temperature of $T = 294$ K. At $Q < 0.1$ Å^{-1}, the thin solid lines indicate the Sadakane theory. At $Q > 0.13$ Å^{-1}, the power law behavior Q^x is shown. All statistical errors are indicated by the vertical lines, mainly visible at the highest Q. **Inset:** the exponent x of the high-Q scattering as a function of temperature. The open symbols indicate the orientation averaged values and the solid symbols indicate the horizontal and vertical sections [the average of the two directions corresponds to the average of the temperature dependent experiment (solid line)]. In addition, the expected value from the scaling of the two critical exponents γ and ν is marked by the cross. Within the error bars, the exponent for the vertical direction and the one from the scaling agree well with the theoretical value of $x = -2.75$ of the two-dimensional Ising behavior (short solid red line).

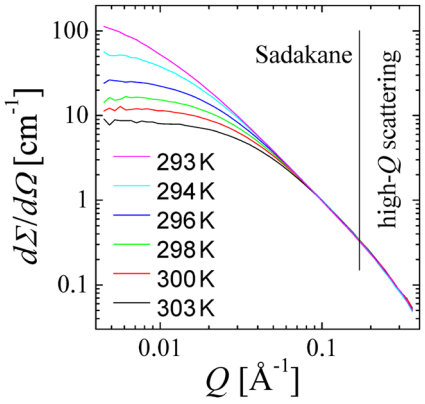

FIG. 4. Orientation averaged macroscopic cross section of the critical fluid as a function of temperature obtained from the SANS experiments. The Q-range for the theory fit of Sadakane *et al.*[6] [Eq. (3)] is indicated by the vertical line. At higher Q, a second range for a power law Q^x is obtained. The statistical errors for $Q > 0.02$ Å^{-1} are similar to the width of the line, but can be seen by the wiggles at smaller Q.

However, the aspect ratio is quite close to unity. While the power law exponent x is closer to −3 in the vertical direction, it is closer to −2 in the horizontal direction. Comparing this with the theoretical part [Eqs. (4) and (5)], the CDW are oriented and propagate in the horizontal direction, while the two-dimensionally confined density fluctuations extend in the remaining directions.

The high-Q exponents x of the observed power law Q^x at $Q > 0.13$ Å^{-1} are given in the inset of Fig. 3. First, the orientation-averaged values $x = -2.38 \pm 0.02$ are practically constant with temperature (see Fig. 4 and following text) and agree with the average exponent obtained for the horizontal and vertical sections at 294 K. It also compares well with the prediction in Eq. (6) of $x = 2.375$ with η_z set to 0. The exponent $x = -2.08 \pm 0.06$ for the horizontal direction is slightly smaller than −2. The integer value of −2 is associated with sharply confined domains [Eq. (5)]. The exponent $x = -2.69 \pm 0.07$ for the vertical direction is slightly greater than −3, which can be associated with fluctuations confined in two dimensions [Eq. (4)] and $\eta_{xy} = 1/4$. Comparing the exponent x from the scattering profiles with the ratio of the critical exponents γ and ν (Table I) according to the scaling relation $\eta_{xy} = 2 - \gamma/\nu$ and taking the integer exponent −3 as given; one obtains the same exponent within the statistical errors, thus confirming $\eta_{xy} = 1/4$. The values $x = -2.08$ and −2.69 of the horizontal and vertical sections, respectively, which differ from the ideal exponents −2 and −2.75, could also be due to the finite acceptance angle of ±15° and the finite magnetic field of 2 T, which deviate from ideal conditions.

TABLE I. Obtained parameters of the critical behavior.

Parameter	Value
T_C (K)	292.30 ± 0.15
S_0 (cm^{-1})	0.280 ± 0.009
γ	1.046 ± 0.007
ξ_0 (Å)	4.74 ± 0.16
ν	0.590 ± 0.007

Figure 4 shows the orientationally averaged scattering profiles of the critical fluid for different temperatures. The magnetic field of 2 T was applied on in the horizontal direction. In comparison with Fig. 3, we see the much reduced statistical noise that provides more reliable parameters for the forward scattering and an average correlation length that then results in a critical behavior (Fig. 5) with very little statistical noise. Going from higher to lower temperatures, the low Q scattering becomes more critical, while at medium $Q \approx 0.07$–0.17 Å^{-1}, the intensity is almost independent of temperature and a Q^{-2} power law is observed. For this Q-range below 0.17 Å^{-1}, the theory of Sadakane[6] [Eq. (3)] can be successfully applied. The agreement of the theory with the measurements is not shown here because it is difficult to distinguish. The value of the Debye length λ_D could be fixed to values of 7 Å or less and did not change the quality of the fit. In addition, the obtained values of κ remained almost constant (as we will see in the following), and in a second fitting cycle, the value was fixed to strengthen the statistical confidence of the forward scattering $S(0)$ and the correlation length ξ. At even higher $Q > 0.17$ Å^{-1}, the influence of the correction by the critical correlation function exponent η becomes visible. Again, the dependence of the scattering profile at high Q is nearly independent of temperature.

The criticality of the forward scattering $S(0)$ obtained from the Sadakane theory fit is shown in Fig. 5. The x axis is spanned by the reduced temperature τ [see Eq. (7)], where T_C is 293.3 ± 0.2 K. The critical exponent $\gamma = 1.05 \pm 0.01$ was found. Astonishingly, it is much closer to the mean field value $\gamma = 1$ compared to a two-dimensional confined case ($\gamma = 7/4 = 1.75$) as found before.[6,10] The exact reasons will be rationalized in the discussion. A quite similar critical behavior of the correlation length ξ is obtained, now with the exponent $\nu = 0.59 \pm 0.01$. Again, this value is much closer to the mean field value $\nu = 1/2$ compared to a previously found value of $\nu = 1$ for two-dimensionally bounded domains.[6,10] The critical behavior parameters are summarized in Table I.

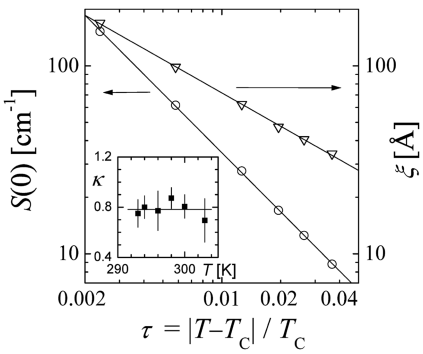

FIG. 5. Experimental forward scattering $S(0)$ (open circles) and the correlation length ξ (open triangles) as a function of the reduced temperature τ. The power law behavior [Eq. (7)] is indicated by the solid line. The error bars are similar to the symbol size. **Inset:** the parameter κ of the Sadakane theory[6] [Eq. (3)] as obtained from the SANS scattering profiles for all temperatures. The horizontal line indicates the mean value.

For completeness, the parameter κ is plotted as a function of temperature in the inset of Fig. 5. It is constant within the error bars.

A quite good orientation of the domains was achieved with the wave fronts of the CDW propagating in the horizontal direction. In addition, all ions are expected to be present in both domains, since the butyric acid is dissociated and thus has a good miscibility with water. In addition, the trivalent thulium ions attract both the negatively charged butyric acid ions and the electron clouds of the water molecules. Therefore, the presence of thulium ions is evenly distributed along the two-dimensional fluctuating domains, and only depletion zones of thulium are found in the regions where the surfactant tails are mixed with butyric acid. Thus, the relevant magnetic structure is extremely extended along the planes of the two-dimensional confined domains and appears thin in the normal, i.e., horizontal, direction. This is directly related to the fact that the magnetic field orientation is along the horizontal direction and agrees with the positive magnetic anisotropy found in the literature.[35] The coordinate system that would explain all the findings is shown in Fig. 2.

This brings us to the enrichment and depletion of either water or butyric acid. This enrichment occurs at the surfactant layers where the counterionic head groups and the hydrophobic tails are present. The mediating trivalent thulium leads to rather average concentrations in the center of the "domains" (Fig. 2). This also explains the mean field-like exponents (Table I): the centers of the domains do not reach the extreme concentrations of either pure water or butyric acid. Thus, the amplitudes of the density fluctuations are significantly reduced, which is directly related to the mean field-like behavior.

The critical correlation function exponents η_{xy} and η_z are related to the interfacial properties of the domains. While $\eta_{xy} = 1/4$ is supported by the SANS profiles in the vertical direction and the scaling relation, the negative value $\eta_z = -0.08 \pm 0.06$ is explained by imperfections of the measurement and $\eta_z = 0$ can be motivated well. In addition, the x and z average exponent $x = -2.38 \pm 0.02$ corresponds very well to the ideal exponent of Eq. (6) ($x = 2.375$) with $\eta_z = 0$. When switching the magnetic field off (supplementary material), we obtain $x = -2.50 \pm 0.02$ that corresponds well to an averaging in three directions, i.e., $x = -8/3 + (2\eta_{xy} + \eta_z)/3 = -5/2$. Comparing this result with a previous measurement[16] without field and an antagonistic salt serving the CDW, we obtained $x = -2.29 \pm 0.03$. Here, it seems that the one-dimensional Ising $\eta_z = 1/2$ is possibly observed as claimed in that publication, although the combination of exponents was motivated differently. Therefore, an ordering of that system with magnetic field is still desired with the challenge of observing a one-dimensional Ising η_z.

Critical fluids with surfactants and ions[6–12] are interesting as model systems for electrolytes[1] and also for fundamental questions about the Ising criticality with different dimensionality.[15] They consist of water and a slightly hydrophobic substance that tend to de-mixing and thus to criticality. The surfactant gives rise to the CDWs that confine the critical fluctuations to two dimensions. The critical exponents γ and ν of a two-dimensional Ising fluid were observed.[6,10] After that, the critical exponent η was discussed separately as it was obtained from the high-Q small-angle scattering.[16] Here, the domains were not aligned, and so only orientationally averaged information could be extracted. However, first ideas about different scattering profiles in different directions were developed. In the current study, we managed to orient the domains successfully. That lead to independently determined exponents $\eta_{xy} = 1/4$ and $\eta_z = 0$ within the experimental precision [the main limitation results from the finite angular section ($\pm 15^\circ$) that is a compromise between statistical noise and representing the exact direction]. Therefore, the existence of a two-dimensionally confined fluid was confirmed and in the last direction a simple lamellar order was observed. The critical fluctuations nearly displayed critical exponents γ and ν of a mean field system. That was explained by the long-range interaction of the many ions in the system.[34,43] It is interesting that still the value $\eta_{xy} = 1/4$ is connected to the two-dimensional Ising criticality. The whole study makes it clear that aligning domains is not trivial and may be accompanied with additional physical mechanisms that initially were unexpected. However, extraction of the critical exponent η tells much about the domain boundaries of the fluctuating fluid. In this sense, it is of fundamental interest for the physics of complex fluids and the Ising criticality in different dimensions.

The supplementary material provides more details about the sample procurement, preparation, and chemistry. Furthermore, details about the SANS instrument and measurements are given. Finally, the separate temperature SANS scan without magnetic field is displayed with the important high-Q power law.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Debasish Saha: Formal analysis (equal); Investigation (lead); Writing – original draft (equal); Writing – review & editing (equal). **Steven Parnell**: Formal analysis (equal); Investigation (equal). **Dirk Honecker**: Investigation (equal). **Eunjoo Shin**: Data curation (equal); Investigation (equal). **Kuno Schwärzer**: Resources (equal). **Stephan Förster**: Funding acquisition (lead); Supervision (equal). **Henrich Frielinghaus**: Conceptualization (lead); Formal analysis (lead); Investigation (equal); Supervision (lead); Writing – original draft (lead); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available in ISIS at https://doi.org/10.5286/ISIS.E.RB2420122 (Ref. 42).

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