Scaling of nonlinear susceptibilities in an artificial permalloy honeycomb lattice

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Two-dimensional artificial magnetic honeycomb lattice is predicted to manifest several magnetic phase transitions as a function of reducing temperature. We have performed the analysis of nonlinear susceptibility to explore the equilibrium nature of phase transition in artificial honeycomb lattice of ultrasmall connected permalloy ($Ni_{0.81}Fe_{0.19}$) elements, typical length of $\simeq$ 12 nm. The nonlinear susceptibility χ_{n1} is found to exhibit an unusual crossover character in both temperature and magnetic field. The higher order susceptibility χ_3 changes from positive to negative as the system traverses through the spin solid phase transition at $T_s = 29$ K. Additionally, the static critical exponents, used to test the scaling of χ_{n1}, do not follow the conventional scaling relation. We conclude that the magnetic phase transition, especially to the low temperature spin solid order, is not conventional in nature at this length scale.

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The interplay between magnetic and thermodynamic characteristics often dictates the nature of phase transition in a magnetic material. Magnetic materials that exhibit equilibrium phase transition, such as spin ice or spin glass, aptly manifest this tendency [1–3]. More recently, artificial magnetic honeycomb lattice has emerged as new venue to explore many equilibrium phenomena of geometrically frustrated magnets in a disorder-free environment [4–8]. The underlying physics in a two-dimensional honeycomb lattice is controlled by the peculiar moment arrangements of "two-in & one-out" (or vice versa) or "all-in or all-out" configurations on a given vertex of the lattice [4,6]. The two-in & one-out refers to a situation where two moments, aligned along the elements of the honeycomb lattice, are pointing towards the vertex and one moment is pointing away from it; also termed as the quasi-ice rule [9]. Theoretical researches have shown that an artificial magnetic honeycomb lattice can undergo a series of thermodynamic phase transitions as a function of reducing temperature from a paramagnetic phase, consisting of the distribution of two-in & one-out (or vice-versa) and all-in or all-out moment arrangements, to a short-range ordered spin ice state [10,11]. For further reduction in temperature, the system tends to develop a magnetic charge ordered state, which is described by the random distribution of chiral vortex loops. At much lower temperature, a honeycomb lattice is predicted to develop a novel ground state of spin solid order, described by the periodic arrangements of the vortex magnetic loops of opposite chiralities [12]. Each magnetic phase transition reduces the overall entropy of the system. The transition to the spin solid ground state is expected to be truly thermodynamic in nature, with zero entropy and magnetization at low temperature [10,13,14].

Analysis of nonlinear susceptibility provides an ideal method to test the equilibrium nature of a magnetic phase transition [15–17]. An equilibrium phase transition is manifested by the scaling of nonlinear susceptibilities where the static critical exponents are related to each other via a conventional relation. To understand the equilibrium nature of the spin solid order, it is desirable to investigate the properties of nonlinear susceptibilities in artificial honeycomb lattice. Previous efforts in accessing the ground state of spin solid order have mostly focused on the disconnected geometry of the honeycomb lattice where thin elements, of length varying between $\simeq$ 500 nm and 2 μm, are separated enough to reduce the interelemental energy of the lattice [18,19]. More recently, we proposed a new sample design to create artificial honeycomb lattice of "connected" ultrasmall permalloy ($Ni_{0.81}Fe_{0.19}$) elements, with a typical element dimension of $\simeq$ 12 nm (length) $\times$ 5 nm (width) $\times$ 7 nm (thickness) [20]. Details about the fabrication procedure can be found elsewhere [21]. At this length scale, the estimated interelemental energy $\simeq$ 12 K is small enough to allow temperature to be a feasible tuning parameter to explore the temperature dependent evolution of magnetic phases, including the spin solid order. Using magnetic, neutron reflectometry and small angle neutron scattering measurements, previously we demonstrated the phase transition to the long-range ordered spin solid state at low temperature $T \leqslant 30$ K in the newly designed honeycomb lattice [20,22]. In this report we show that the development of spin solid state is accompanied by a change in the nature of nonlinear correction to the linear susceptibility χ_1. As the system traverses through the spin solid transition at $T_s \simeq 29$ K, the nonlinear term χ_3 changes from negative to positive, which is atypical of magnetic phase transition. Also, a crossover between low field and high field regimes is detected, which leads to two different scaling analysis of nonlinear susceptibilities. The estimated static critical exponents do not follow the conventional scaling relation. Together, these phenomena suggest that the transition to the spin solid state is not truly equilibrium in nature in artificial honeycomb lattice of connected ultrasmall elements.

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In the case of an equilibrium phenomenon, the nonlinear susceptibilities exhibit a scaling behavior according to the single parameter, given by [3,17,23]

$$\chi_{n1}(T,H) = H^{2/\delta} f(\tau^{(\gamma+\beta)/2}/H), \quad (1)$$

where $\tau = (T/T_s) - 1$, γ is the static critical exponent describing the divergent nature of magnetic susceptibility as a function of temperature, and β is the magnetic order parameter critical exponent. The determination of nonlinear susceptibility χ_{n1} plays the key role in this exercise. The nonlinear susceptibilities are written as the higher order terms in following equations [17,23]:

$$M/H(T) = \chi_1(T) - \chi_3(T)H^2 + O(H^4) \quad (2)$$

$$= \chi_1(T) - a_3(T)\chi_1{}^3 H^2 + O(H^4) \quad (3)$$

$$\chi_{n1}(T,H) = 1 - M(T,H)/\chi_1 H. \quad (4)$$

where $\chi_1(T)$ is the linear susceptibility at temperature T, $\chi_3(T)$ is the nonlinear susceptibility, coefficient $a_3 = \chi_3/(\chi_1)^3$, and χ_{n1} is the net nonlinear susceptibility.

Determination of the critical exponents γ and β depends on the asymptotic nature of the arbitrary scaling function $f(x)$, with the boundary conditions $f(x) = \text{const}$ as $x \to 0$ and $f(x) = x^{-2\gamma/(\gamma+\beta)}$ as $x \to \infty$. The nonlinear susceptibility $\chi_{n1}(T,H)$ is expected to follow power-law dependence in both T and H with two independent static critical exponents γ and δ, respectively. The power-law dependencies are described by the following expressions [3,17]:

$$\chi_{n1}(T) \propto \tau^{|\gamma|}, \quad (5)$$

$$\chi_{n1}(T \simeq T_s, H) \propto H^{2/\delta}. \quad (6)$$

The two independent exponents γ and δ are related to the magnetic order parameter critical exponent β via the following scaling relation:

$$|\delta| = 1 + |\gamma/\beta|. \quad (7)$$

The above scaling relation represents a robust test, arguably, of the true equilibrium phase transition in a magnetic system. Magnetization data on the newly designed artificial permalloy ($Ni_{0.81}Fe_{0.19}$) honeycomb lattice were obtained in the field range of 10–1500 Oe using a commercial magnetometer. The sample was slowly cooled from $T = 350$ K to the desired temperature before collecting the data. Extra care was taken in removing magnetic hysteresis in the superconducting magnet of the magnetometer by cycling the magnetic field in oscillatory mode several times at $T = 350$ K before cooling to the measurement temperature. At each field, the system was allowed to sufficiently relax before collecting the data. In Fig. 1 we plot the M vs H data at a few characteristic temperatures. The total magnetization at higher temperature is stronger at low field. The trend reverses across the crossover field, which also varies with temperature. The linear susceptibility $\chi_1(T)$ at different temperatures were determined by fitting the M versus H curves at low fields, see Fig. S1 in the Supplemental Material [24]. We have analyzed the first and second order term in the magnetization data. Beyond the second order term, the nonlinear susceptibility becomes much smaller to be of any quantitative importance. Therefore, Eq. (2) reduces to $\chi_3(T,H)H^2 = 1 - M(T,H)/\chi_1 H$. Hence, $\chi_{n1}(T,H)$ becomes $(\chi_3/\chi_1)(T,H)H^2$ [25].

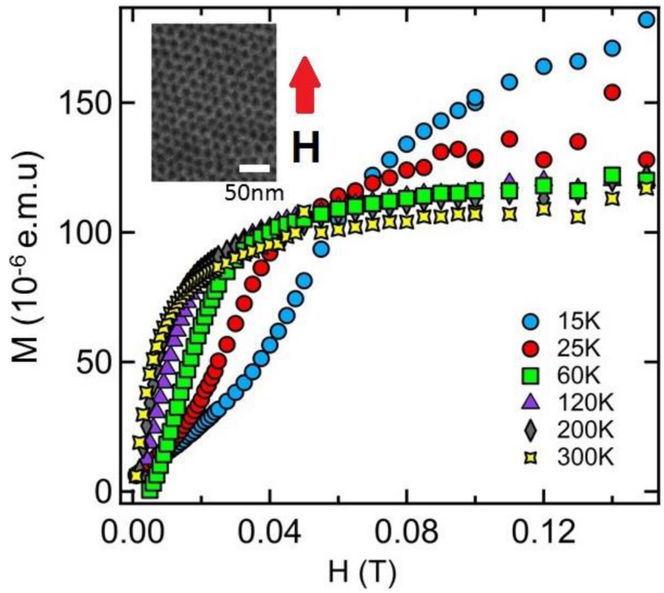

FIG. 1. Magnetization as a function of field. Here magnetization is plotted as a function of field at different temperatures. Magnetization data exhibits a crossover behavior in field. While the higher temperature susceptibility is stronger at low field, the magnetization at low temperature is larger above the crossover field $H \simeq 0.04$–0.06 T. Inset shows the scanning electron micrograph of a typical artificial honeycomb lattice of ultrasmall elements. Magnetic field was applied in-plane to the lattice.

In Fig. 2(a) we have plotted net nonlinear susceptibilities $\chi_{n1}(T,H)$ as a function of H^2 at different temperatures between $T = 10$ and 300 K. The plot of nonlinear susceptibility reveals several very interesting behaviors in applied field. First, at low temperature, $T \leqslant 25$ K, χ_{n1} is negative for the entire field application range. The negative nonlinear susceptibility suggests that the higher order correction to the linear susceptibility is very strong. Surprisingly, negative χ_{n1} is only observed below the spin solid phase transition. Second, the nonlinear susceptibility not only becomes positive above $T \simeq 30$ K, but also exhibits an unusual trend at low field. At low field, χ_{n1} first decreases before manifesting a gradual enhancement as the applied field strength increases. Thus, the slope of the curve changes from negative (regime 1) in low field to positive (regime 2) in high field. Additionally, the slope of the curve also changes as a function of temperature at low field: from positive at $T \leqslant 30$ K to negative at $T \geqslant 30$ K. We summarize these observations in plot of χ_3 vs T in different field regimes in Figs. 2(b) and 2(c). In general, nonlinear correction to the susceptibility only changes in magnitude, not in sign. This is a puzzling behavior in artificial honeycomb lattice. The characteristic crossover field, separating the two distinct regimes, decreases as the measurement temperature increases [see inset in Fig. 2(a)]. We also notice that the saturated value of χ_{n1} increases as temperature increases. The net magnetization is expected to decrease as temperature reduces in artificial honeycomb lattice. First, we analyze the nonlinear susceptibility data above the characteristic field (in

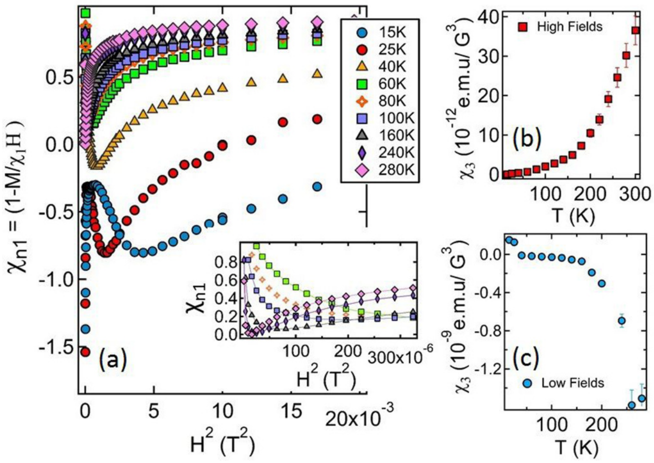

FIG. 2. Nonlinear susceptibility χ_{n1} as functions of field and temperature. (a) χ_{n1} is estimated using Eqs. (2)–(4) where χ_1 is obtained from fitting M vs H plot at low field. Two features are immediately obvious in this figure: a change in the sign of overall nonlinear susceptibility across $T \simeq 30$ K and a crossover regime in field and temperature. As shown in the inset of the figure, the slope of the curve changes from negative to positive at some field value. We call it characteristic crossover field, which increases as temperature decreases. (b) and (c) Higher order susceptibility χ_3 as a function of temperature across the crossover field. χ_3 increases as a function of temperature in high field regime 2 (b) and becomes more negative in low field regime 1 (c).

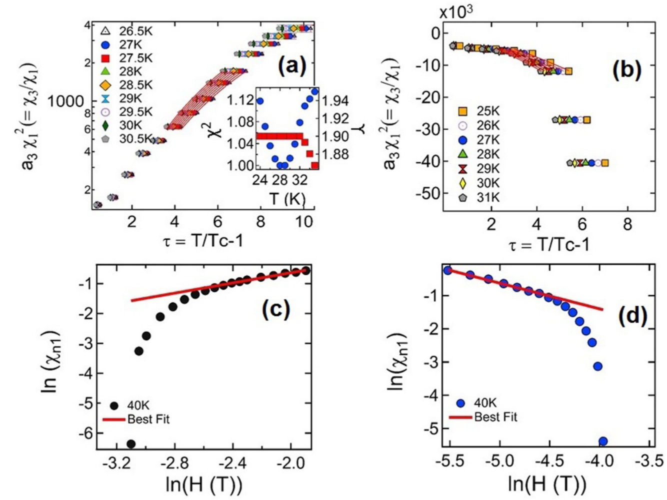

FIG. 3. Estimation of static critical exponents γ and δ. (a) To estimate the critical exponent γ, the coefficient a_3 (see text for detail) is plotted as a function of $\tau = (T/T_s) - 1$ for different T_s values, across the spin solid transition at $T = 30$ K. γ is estimated by fitting the fixed number of points in the divergence regime of the curve using Eq. (5). Best fit to the experimental data is obtained for the critical exponent $|\gamma| = 1.9$ (inset shows the plot of fitting parameter χ^2 vs γ). (b) Similar analysis is performed for the low field regime 1, with estimated $|\gamma| = 1.4$. In both regimes, best fit corresponds to spin solid transition at $T_s = 29$ K. Nonlinear susceptibility χ_{n1} is plotted as a function of field at temperature near T_s. Experimental data are fitted using the asymptotic function in Eq. (6) to obtain critical exponent $|\delta|$ (c) in high field regime 2, $\simeq 2.4$ and (d) low field regime 1, $\simeq 2.5$.

regime 2). Even in regime 2, the maximum value of the field, up to which $\chi_{n1}(T)$ is linear in H^2, decreases gradually as T approaches T_s. It suggests that the higher order corrections in the net susceptibility is still significant [25]. The linear portion of $\chi_{n1}(T)$ at different temperatures are fitted with Eq. (3) to extract the coefficient $a_3(T)$.

To verify the equilibrium nature of magnetic phase transition to the spin solid state, first we extract the exponent γ using the formalism, described above, in Eq. (5). For this purpose, the nonlinear susceptibility $\chi_{n1} = a_3\chi_1^2$ is plotted as a function of τ for few different choices of spin solid transition temperatures $T_s \in [25,35]$ K in Fig. 3(a). We have fitted a fixed number of data points, in the divergence regime, on each curve using Eq. (5). Estimated γ is found to vary in the range of [1.7, 2]. The best fit is obtained for $T_s = 29$ K, with the corresponding value of $|\gamma| = 1.9$ [see inset in Fig. 3(a)]. The transition temperature T_s is very close to the experimental value of $T = 30$ K, as estimated from the previous dc susceptibility and electrical measurements [20,21]. Also, the static critical exponent γ is comparable to the value ($|\gamma| \simeq 2.25$) found in systems manifesting truly thermodynamic phase transition, such as interacting arrays of nanoislands or spin freezing in canonical and geometrically frustrated systems [3,15,17,23,26]. Similar analysis was performed in the low field regime (regime 1) below the characteristic crossover field. The best fit is obtained for the static critical exponent $|\gamma| = 1.4$, see Fig. 3(b). It is not very different from the magnitude of γ in the high field regime (regime 2). It seems that the crossover phenomenon, manifested by the change in the slope of $\chi_{n1}(T)$ as the system traverses across the transition temperature at a given field, does not affect the estimation of γ and the transition temperature T_s in the honeycomb lattice of ultrasmall elements.

Next, we determine another critical exponent δ by plotting $\ln(\chi_{n1})$ versus $\ln(H)$ at temperature near the spin solid transition. The experimental data is fitted using the asymptotic function in Eq. (6). As shown in Fig. 3(c), a good fit to the data is obtained for the critical exponent $\delta = 2.4$ in regime 2. Similar analysis in regime 1 at low field yields $|\delta| = 2.5$, which is also similar in magnitude as found in the high field regime 2. Finally, we test the scaling behavior of nonlinear susceptibilities, as described by Eq. (1). If the magnetic phase transition to the spin solid state in artificial honeycomb lattice is indeed a true equilibrium phase transition, then the nonlinear susceptibilities should exhibit the scaling behavior due to the estimated critical exponents. According to Eq. (7), for critical coefficients $|\gamma| = 1.9$ and $\delta = 2.4$, the magnetic order parameter critical exponent β is $\simeq 1.4$. As shown in Fig. S2 in the Supplemental Material [24], the nonlinear susceptibilities at different temperatures do not exhibit the scaling collapse on one curve for the estimated exponents. To explore the scaling behavior further, we vary the critical exponents γ, δ, and β systematically. First we discuss the scaling in regime 2. A scaling behavior is observed for exponents $\delta = 10$ and $|\gamma| = 1.5$, see Fig. 4(a). Although exponent $|\gamma|$ is similar to the estimated value, scaling collapse of χ_{n1} data only occurs for δ much larger than the estimated value. At large x values, some data scatter from the scaling curve due to the large errors associated with the smaller nonlinear susceptibilities.

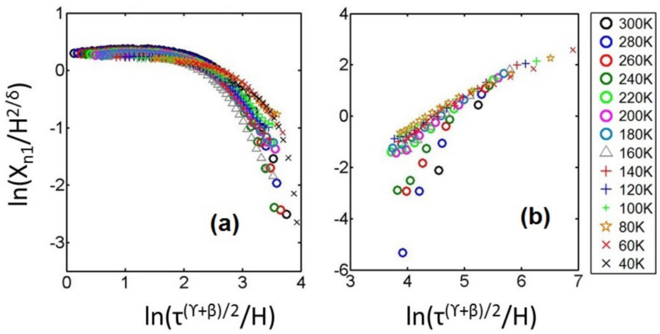

FIG. 4. Scaling analysis of nonlinear susceptibilities in artificial honeycomb lattice. (a) Nonlinear susceptibilities exhibit scaling behavior for $|\gamma| = 1.5$, $\delta = 10$, and $|\beta| = 0.1$. The critical scaling coefficients do not satisfy the scaling relation in Eq. (7). (b) Similar analysis was performed in the low field regime 1. Interestingly, the nonlinear susceptibilities exhibit scaling behavior for the same set of critical exponents, as in high field regime 2.

We also tested the scaling behavior for intermediate values of δ, 4.75, while keeping the coefficient γ constant. The scaling of nonlinear susceptibilities improves as δ increases. However, the critical exponents do no follow the scaling relation, outlined in Eq. (7).

The scaling behavior was also tested for nonlinear susceptibilities in low field regime 1. For uniformity, we have used the estimated static critical exponents of $|\gamma| = 1.4$, $\delta = 2.5$, and $|\beta| = 0.95$ for the scaling analysis. As shown in Fig. S3 of the Supplemental Material, the nonlinear susceptibilities do not scale for the calculated values of exponents. To our surprise, χ_{n1} data at different temperature exhibit scaling characteristic for the similar set of exponents, $|\gamma| = 1.4$, $\delta = 10$, and $|\beta| = 0.1$, that are used to obtain scaling collapse in the high field regime 2, see Fig. 4(b). Once again, the critical exponents do not satisfy the scaling relation in Eq. (7). It further confirms that the magnetic phase transition to the spin solid state is not thermodynamic in nature. The observed consistencies in the estimation of critical exponents as well as in the scaling analysis in two different regimes of χ_{n1} constitute a unique aspect of the spin solid phase transition. It suggests that the nonlinear correction to magnetic susceptibility in spin solid phase is subtly similar to that in the high temperature phases. The discrepancies between the estimated values of the static critical exponents and that used for the scaling manifestation can be attributed, arguably, to the formation of small ferromagnetic clusters with short-range order at intermediate temperatures, which ultimately enhances χ_{n1} considerably and led to strong but noncritical background temperature dependence. Similar behavior was previously observed in magnetic systems that exhibit nonequilibrium phase transition [27].

Our investigation of the equilibrium nature of magnetic phase transition in artificial honeycomb lattice has revealed two important properties that are not conventional in nature: first, the nonlinear susceptibility exhibits a crossover behavior in both temperature and magnetic field. The slope of χ_{n1}, which is used to determine the strength of the nonlinear correction to the overall magnetic susceptibility, is found to change from negative, at low field, to positive, at high field. Also, the net nonlinear susceptibility χ_{n1} changes from positive to negative in temperature. This crossover occurs across the spin solid phase transition temperature at $T \simeq 30$ K. A magnetic phase transition is not known to depict such contrasting characteristic across the transition temperature. Clearly, the underlying magnetism in artificial honeycomb lattice does not fit congruently with the conventional understanding. Second, the experimental data do not exhibit scaling behavior for the estimated values of critical exponents. Rather, a scaling collapse of χ_{n1} requires much larger value of the critical exponent δ; not typically observed in a magnetic material with equilibrium phase transition. Also, the static critical exponents do not satisfy the conventional thermodynamic scaling relation. The overall scaling behavior suggests a nonconventional nature of the transition, which can be arising either due to the finite spin dynamics in the system or, a distribution of relaxation times in short-range ordered magnetic clusters, such as spin ice order or the vortex loop type magnetic correlation across one honeycomb. A distribution of relaxation times in magnetic clusters is known to cause nonconventional scaling behavior. The presence of spin dynamics or the distribution in spin relaxation rate, especially at low temperature, will result in finite entropy accumulation. It is worth pointing out that the large element size honeycomb lattice, with much larger dipolar interaction energy, may exhibit different nonlinear magnetic response. Further research works are highly desirable to fully understand the perplexing observations reported here as well as to explore the implication to large element size honeycomb lattice, especially in the disconnected structure.

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