Possible multipolar density wave in NdFe$_2$Ga$_8$

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We study the magnetism in NdFe$_2$Ga$_8$ by the neutron-diffraction and thermal-expansion techniques. Thermodynamical measurements have demonstrated that there are two magnetic transitions at 10 and 14.5 K, respectively. Neutron-diffraction measurements confirm that the lower one is an antiferromagnetic transition with a commensurate magnetic structure. Both the commensurate and incommensurate magnetic peaks are found below the higher transition but their intensities only gradually increases with decreasing temperature. Below 10 K, the commensurate peak intensity increases quickly with decreasing temperature, signaling the antiferromagnetic transition, while the incommensurate peak intensity disappears below 5 K. We attribute the high-temperature magnetic transition as a multipolar order, which induces incommensurate magnetic peaks. The multipolar ordering is suppressed by field at about 7 T, where the linear Grüneisen parameter along $c$-axis diverges with decreasing temperature as $T^{-1}$. Our results suggest that NdFe$_2$Ga$_8$ exhibits a multipolar density wave that is analogous to the spin density wave for the dipole moment.

I. INTRODUCTION

A multipolar order (MO) happens rarely in magnets as the interactions between dipole moments are typically much stronger than those between multipole moments. There are increasing number of $f$-electrons compounds that exhibit MOs due to the interplay between spin and unquenched orbital degrees of freedom [1, 2]. In many cases, MOs have been treated as hidden orders, since the typical symmetry-breaking probes cannot directly observe them. Indeed, the famous hidden order in URu$_2$Si$_2$ has been proposed to originate from the MOs [3-7]. As magnetic orders from dipole moments, e.g., the antiferromagnetic (AFM) and ferromagnetic (FM) orders, the MOs can have both thermal and quantum transitions and exhibit many interesting properties [8-11]. The analysis of the MO is traditionally based on the local point group symmetry and the crystal-electric field (CEF). This single-ion approach works well if the $f$ electrons are localized, i.e., the atomic $f$-orbitals are good enough to describe the MO. However, one has to also consider the hybridization between the $f$ and conduction electrons and the strong electron correlation effect to correctly capture the multipolar physics in many materials. For example, the itinerancy of electrons has been suggested to play a crucial role for the MO in CeB$_6$ [12, 13] and possibly in URu$_2$Si$_2$ [5, 7]. However, the itinerant MOs in these materials are not the simple density-wave type since the local moments still play important roles.

The material studied here is NdFe$_2$Ga$_8$ (space group $Pbma$) with Nd$^{3+}$ ions form one-dimensional (1D) chains along the $c$ axis. It belongs to the so-called 1-2-8 systems, i.e. $LM_2T_8$ ($L =$ La, Nd, Ce, etc., $M =$ Fe, Co, Ru, etc. and $T =$ Al, Ga, In), which have shown interesting magnetic properties from 4$f$ electrons [14-28]. Previous studies have shown that NdFe$_2$Ga$_8$ has two magnetic transitions at zero field [29]. Both transitions are hardly affected by the magnetic field within the $ab$ plane, but they can be quickly suppressed by the field along the $c$-axis at 7 T, showing three-dimensional (3D) spin-density-wave (SDW) type of quantum criticality [29]. While both transitions have been assumed to be AFM in nature, the magnetic structures have not been identified.

In this work, we further studied the magnetic structure of NdFe$_2$Ga$_8$ by the neutron-diffraction technique. We found that the transition at $T_N \sim 10$ K is from a long-range commensurate AFM order. Below the higher transition temperature $T_D \sim 14.5$ K, both incommensurate (IC) and commensurate magnetic peaks gradually appear. The intensities of the IC peaks become maximal at $T_N$ and decrease to zero at about 5 K. Interestingly, the linear Grüneisen parameter along $c$-axis shows $T^{-1}$ divergent behavior at low temperatures at about 7 T, suggesting the existence of a quantum critical point (QCP) [30, 31]. Our results suggest that the magnetic transition at $T_D$ is associated with a MO, which may be a multipolar density wave (MDW) that can be analogous to the spin density wave (SDW) for the dipole moment.
II. EXPERIMENTS

Single crystals of NdFe$_2$Ga$_8$ were grown by the self-flux method as reported previously [29]. Neutron single-crystal diffraction (NSCD) measurement was carried out for a 113-mg single-crystal sample on TOPAZ diffractometer at the Spallation Neutron Source, Oak Ridge National Laboratory. Five grams of single crystals were ground to powders and measured by the neutron powder diffraction (NPD) on the Echidna diffractometer at the Australian Nuclear Science and Technology Organization. The refinements for the NSCD and NPD data were done by using the Jana and Fullprof programs, respectively [33, 34]. The linear Grüneisen parameter was measured by a home-made temperature-modulated dilatometer [35] putting in a physical property measurement system (Quantum Design) and a 18-T magnet with a top-loading $^3$He fridge.

III. RESULTS AND DISCUSSIONS

Figure 1(a) and 1(b) show NSCD patterns in the (0,K,L) plane at 5 and 20 K, respectively. Compared to 20-K data, new peaks appear at 5 K at half integer L, suggesting the appearance of long-range AFM order. Nuclear structure was refined at 20 K with 23033 peaks and the result is the same as previously reported [29]. At 5 K, no change of nuclear structure is found and the magnetic structure is refined by the 5231 magnetic peaks with a total wR of 26.36. Figure 1(c) shows the NPD data at 3 K and the refinement for both nuclear and magnetic structures gives the magnetic R-factor as 60.5. About 3.2% of FeGa$_3$ impurity is found in the NPD pattern. We note that this impurity does not affect the NSCD refinement as it has the form of polycrystalline and its diffraction intensity in the single-crystal sample is thus very weak. In both refinements, the magnetic form factor has included both $< f_0 >$ and $< f_2 >$ terms. Both the single-crystal and polycrystalline refinements give the same magnetic structure as shown in the inset of Fig. 1(c). The magnetic moments point to the c-axis, and are aligned ferromagnetically within the ab plane but antiferromagnetically along the c axis. The value of the ordered moment from the NSCD and NPD is 3.379 ± 0.016 $\mu_B$ at 5 K and 3.31 ± 0.15 $\mu_B$ at 3 K, respectively.

We note that the above refinements have assumed that the moments are on the Nd sites. If the moments are added on Fe sites, the value on Fe1 site is negligible while that on Fe2 site is about 0.593(29) and 0.544(27) $\mu_B$ for Fe$^{2+}$ and Fe$^{3+}$ magnetic form factors used in refinements, respectively, which are much smaller than the moments on Nd sites. However, we find no improvement to the fit with the Fe moments included and the Nd moments are almost the same as before. Therefore, we conclude that Nd$^{3+}$ is the only magnetic ion in the magnetic ordering of NdFe$_2$Ga$_8$. Similar conclusion has also been found in PrFe$_2$Al$_8$ [24].

Figure 2(a) and 2(b) show the magnetic Bragg peaks at (1,4,0.5) cutting along the H and K directions, respectively. With the peak intensity decreasing with the increasing temperature, the peak can always be well fitted by the Gaussian function with no change of the width. For the cuts along the L direction (Fig. 2(c)), while the peak can still be fitted by the Gaussian function at low temperatures, two new peaks emerge at the IC positions ($L \sim 0.5 \pm \delta$) with $\delta \approx 0.05$ at high temperatures. A three-Gaussian function with the sum of three individual Gaussian functions is thus introduced to fit the data, as shown in Fig. 2(d).

Figure 2(c) shows the colormap for the intensity as a function of L and temperature. The incomprehensibility $\delta$ varies little above 10 K and seems to decrease with decreasing temperature below 10 K. The full-width-at-half-maximums (FWHMs) for the commensurate and IC peaks are both temperature independent and comparable to the instrumental resolution, indicating that both of the peaks are associated with long-range orders.
Below 10 K, the former one can be fitted by the function
peak intensity at the commensurate and IC positions.

The black solid line is fitted by the function as described in
pendence of the peak intensity at (1,4,-0.5) and (1,4,-0.45).

Gaussian function. (d) shows the data in (c) for temperatures
different temperatures. The solid lines are fitted by the three-
L

Figure 2(f) shows the temperature dependence of the linear Gr"uneisen
parameter $\Gamma_c$ at various fields. The measurements were made
in the FC process. The arrows indicate $T_D$ and $T_N$ at zero
field. (b) Field dependence of $\Gamma_c$ at 1.8 K. The arrows indicate
the field-decreasing and field-increasing processes. (c) Field
dependence of $\Gamma_c$ below 1.8 K for a different sample. The
measurements were made with the field-decreasing process.
(d) $\Gamma_c$ as a function of temperature at 7 T in the log-log scale.
The values below 1.8 K are taken from the data in (c) and
normalized to those above 1.8 K. The dashed line represents the $T^{-1}$
dependence.

Figure 2(f) shows the temperature dependence of the peak intensity at
the commensurate and IC positions. Below 10 K, the former one can be fitted by the function
$M_0(1 - T/T_N)^{\beta}$ with $\beta$ and $T_N$ fixed at 0.5 and 10 K,
respectively. The IC peak intensity shows totally different behavior. Above $T_N$, the temperature dependence of both commensurate and IC peak intensities are the same. Within our resolution, we can only identify the appearance of magnetic peaks at 13 K. With decreasing temperature, the IC peak intensity reaches a maximum at $T_N$ and decreases with further cooling temperature, whereas the commensurate peak intensity quickly increases as a regular AFM transition.

To further investigate the nature of the magnetic transitions in NdFe$_2$Ga$_6$, we studied the linear Gruneisen parameter $\Gamma_c$, where the linear thermal expansion is measured along the $c$ axis [35, 36]. Figure 3(a) shows the temperature dependence of $\Gamma_c$ in the field-cooling (FC) process, which is small and positive at high temperatures for all fields. At zero field, $\Gamma_c$ changes to negative with decreasing temperature and its absolute value quickly increases until $T = T_D$. With a kink at $T_D$, $|\Gamma_c|$ continues to increase with decreasing temperature and shows another kink at $T_N$. With field increasing, $T_D$ decreases and seems to merge together with $T_N$ above 3 T, which is consistent with previous reports [29]. Further increasing the field makes it hard to identify both transitions, while $|\Gamma_c|$ at 1.8 K becomes maximum at 5 T and starts decreasing with increasing field. Interestingly, $\Gamma_c$ at low temperature changes continuously from negative to positive value when the field increases from 6 to 7 T.

Figure 3(b) shows the field dependence of $\Gamma_c$ at 1.8 K, where the positive and negative maximums appear at about 7 and 5 T, respectively. Hysteresis behavior is found between increasing and decreasing field processes, which is consistent with previous resistivity and magnetic-susceptibility measurements [29]. The hysteresis is absent at zero field, becomes maximum at about 5 T and finally disappears at about 7 T. Further decreasing temperature makes the maximums at 7 and 5 T more prominent, as shown in Fig. 3(c). Interestingly, two-peak features appear around 5 T at 0.4 K, which indicates complicated behaviors within the ordering state. We note that the transitions at $T_D$ and $T_N$ behave very differently in the field in that the former is easily sup-
pressed by the field whereas the latter is little affected by the field below 3 T where the two transitions merge together [29]. The features around 5 T may thus be related to the couplings between the orders associated with $T_O$ and $T_N$. Nevertheless, here we are more interested in the properties around 7 T, which has been shown to exhibit quantum criticality [29]. Figure 3(d) shows the temperature dependence of $\Gamma_c$ at 7 T, which shows a divergent behavior with $\Gamma_c \propto T^{-1}$ below 2 K.

With the results above, we can get a more comprehensive understanding on the magnetic orderings in NdFe$_2$Ga$_8$. The intensities of the magnetic peaks below $T_O$ increases slowly with decreasing temperature (Fig. 2(d)), which makes it impossible to describe the temperature dependence of the intensities as a conventional AFM order. Therefore, the high-temperature transition at $T_O$ is just like the multipolar transitions in many systems [1, 2] in that the magnetic transition can be clearly revealed by thermodynamical measurements but not neutron diffraction, which is only sensitive to dipole moments. We note that the IC magnetic peaks have also been seen in the MO systems due to the coupling between the multipolar and dipole moments [37–39].

The MO for the transition at $T_O$ explains many strange behaviors of NdFe$_2$Ga$_8$. First, the transition at $T_N$ is most likely a conventional AFM transition with the simple magnetic structure, but this structure cannot be understood by the superexchanges. We note that the Curie-Weiss temperatures for the field within the ab plane and the c axis are -75.8 and 20.6 K, respectively [29]. Naively, one expects that the moments should be aligned antiferromagnetically within the ab plane and ferromagnetically along the c axis. This is totally different from the actual magnetic structure in this system as shown in the inset of Fig. 1(c). This contradiction would be explained if the magnetic structure is strongly influenced by the interactions between multipolar moments. Second, the field only has significant effects for $H//c$. This kind of large field anisotropy has also been widely observed in the MOs in other systems [10–12]. Third, the field dependence of many properties shows a hysteresis behavior. This hysteresis cannot be explained by the FM canting of the moments as it is absent at zero field. This may be understood if we consider that the MO and AFM order are coupled as shown by the temperature dependence of the magnetic peaks and the field dependence of the transitions.

The observation of the MO in NdFe$_2$Ga$_8$ is an unexpected result since the local symmetry of Nd is very low. Surrounded by Ga atoms with nine different positions, the only local symmetry of Nd is the mirror symmetry. In this case, the conventional analysis for the MO based on the local symmetry and CEF does not apply [1, 2]. On the other hand, the quick upturn of the resistivity at $T_O$ [29] and the IC magnetic peaks suggest that the MO should be associated with the itinerant electrons. Especially, the upturn of the resistivity strongly suggests the opening of a gap, which has been widely found for the SDW systems. We thus propose that the MO in NdFe$_2$Ga$_8$ is a MDW, which bears a resemblance to the SDW where the local moments play minor roles. Indeed, we can see that the $T^{-\gamma}$ dependence of the resistivity and the $T^{-1}$ dependence of $\Gamma_c$ at 7 T, which have been traditionally attributed to a SDW QCP [13–14]. We should be cautious though since the QCP in NdFe$_2$Ga$_8$ may involve local moments as $T_N$ and $T_O$ merge together above 3 T. Nevertheless, the sharp upturn of the resistivity at $T_O$ has not been seen in the itinerant MOs studied previously [6, 7, 12, 13], which suggests that NdFe$_2$Ga$_8$ may be a good model system for studying the MDW that is similar to the SDW. It thus provides us an opportunity to study the itinerant MOs from the weak-coupling picture that is supposed to be easier to understand before pursuing the underlying physics of the MOs in more complicated systems.

IV. CONCLUSIONS

In conclusions, we have found both commensurate and incommensurate magnetic peaks in the magnetic ordered states in NdFe$_2$Ga$_8$. While the low-temperature commensurate peaks are associated with the AFM transition at 10 K, the incommensurate peaks cannot be simply by an AFM transition. Considering the crystal structure and relevant experimental results for NdFe$_2$Ga$_8$, we suggest that the magnetic transition at 14.5 K is a multipolar density wave. As the local point group symmetry may play little role here, NdFe$_2$Ga$_8$ could be a good model material to study the itinerant MOs.

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