Two-dimensional Magnetism in κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl, a Spin-1/2 Heisenberg Antiferromagnet with Dzyaloshinskii–Moriya Interaction

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The finite phase transition temperature, T_N , of quasi two-dimensional (2D) Heisenberg antiferromagnetic (AF) crystals results from in-plane anisotropy or coupling between layers. It is usually not known which is the determining factor when both are weak. We show by an electron spin resonance experiment that in the quasi 2D antiferromagnetic crystal, κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl, T_N is determined by in-plane anisotropy while interlayer coupling plays a minor role. The compound has a large isotropic Heisenberg exchange interaction between sites with S = 1/2 spins. The Dzyaloshinskii–Moriya (DM) interaction is the main source of anisotropy, while in-plane anisotropy and the interlayer coupling are very weak. The external-field field-induced static and fluctuating AF magnetizations are independent in adjacent layers above the (zero-field) ordering temperature.

1. Introduction

Magnetic ordering in a two-dimensional (2D) lattice of atoms or molecules is a delicate question.¹⁾ Although theory predicts for most 2D magnets a phase transition at finite temperature T_N , the textbook 2D Heisenberg antiferromagnet (or ferromagnet) with an isotropic interaction between the magnetic sites has a continuous symmetry and does not order at finite temperatures. An anisotropy breaking the continuous symmetry renders T_N finite if the spin on sites is larger than 1/2. Whether ordering occurs at finite temperatures or not in the most interesting S = 1/2 systems depends on the strength of the anisotropy.²⁾

There are few if any experimental studies of truly 2D magnetic ordering; i.e., in systems where the ordering temperature depends mainly on interactions within a single atomic or molecular layer. Crystals with apparently well isolated magnetically ordered layers are found in nature, e.g., YBa₂Cu₃O₆, the antiferromagnetic parent compound of the high temperature cuprate superconductor family, but usually it is not known whether the ordering transition in these is "two dimensional" (in the sense of driven by the anisotropy of interactions within the layer) or "three dimensional", driven by the leftover interactions between layers. The unique properties of the layered crystal of the present experiments allow to test whether magnetic order develops independently in adjacent layers. The aim of this paper is to show that in an organic layered magnet, κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl (κ -ET₂-Cl), the ordering temperature, $T_{\rm N}$ is mainly determined by the in-plane anisotropy and in this sense the crystal is a stack of independent 2D magnets.

2. Principle of the Experiment

One can experimentally distinguish in some layered compounds between ordering driven by intralayer anisotropy or by interlayer coupling (Fig. 1). First consider two idealized 2D single layer models, Type I and II. The isotropic Heisenberg exchange and the Dzyaloshinskii– Moriya (DM) antisymmetric exchange anisotropy are the dominant interactions in both. In the Type I 2D layer there is no other interaction. It has a continuous symmetry in the



Fig. 1. (Color online) Magnetic order as a function of temperature in model layers and crystals. Arrows symbolize order. F_0 , the sum of the isotropic Heisenberg exchange and Dzyaloshinskii–Moriya (DM) interactions is dominant in all systems. In the single layers the intralayer anisotropy is zero in a), finite in b). In the crystals a finite, ordering temperature between weak ferromagnetic and paramagnetic states, T_N , results from (c) interlayer coupling (Type I) or from (d) intralayer anisotropy (Type II). (e, f): In appropriate magnetic fields **H**, the width of the crossover ΔT_{co} is large in all layers of Type I crystals. In Type II crystals ΔT_{co} remains narrow in *A* and broad and shifted to higher temperatures in *B* layers.

plane perpendicular to the DM vector, **D**, and it does not order at finite temperatures. The Type II 2D layer has an additional anisotropic interaction between sites breaking the continuous symmetry and the ordering Néel temperature T_N is finite.

Next consider a Type I 3D crystal, i.e., a stack of weakly interacting 2D Type I layers with a continuous symmetry. The interaction between layers raises $T_{\rm N}$ in the crystal to a finite temperature. Similarly, a Type II crystal is a stack of Type II layers with intralayer anisotropy; but here we assume layers are not interacting. This idealized Type II crystal has the same properties and the same $T_{\rm N}$ as a single Type II layer. The task is to distinguish a crystal of noninteracting Type I layers from a Type II crystal where T_N arises respectively from interlayer coupling or intralayer anisotropy. Type I and II crystals are idealized; in real crystals the question is which interaction is more important in determining $T_{\rm N}$. This can be answered experimentally for a crystal of alternating, chemically equivalent layers, A and B, which have different, symmetry related structures so that the respective DM vectors, \mathbf{D}_A and \mathbf{D}_B are oriented differently. For simplicity we take \mathbf{D}_A perpendicular to \mathbf{D}_B .

Type I and II crystals are distinguished by their different response to an external magnetic field, **H**. The magnetic field transforms the ordering transition into a crossover that depends on the orientation of **H** with respect to **D**. In single layers, a field **H** || **D** has a negligible effect on the width of the crossover if it is not too large. In contrast, if **H** \perp **D** the DM and Zeeman interactions drastically affect the transition. Separately the DM and Zeeman interactions do not induce any antiferromagnetism. However, together they act as an effective spatially oscillating magnetic field³ with the antiferromagnetic wave vector of the ordered state and induce a significant antiferromagnetic order extending well above $T_{\rm N}$.

In Type II crystals in a magnetic field $\mathbf{H} \parallel \mathbf{D}_A$ and $\mathbf{H} \perp \mathbf{D}_B$, *A* layers with a narrow crossover at T_N alternate with *B* layers with a broad crossover extending to much higher temperatures. Type I crystals respond differently to the same magnetic fields. The crossover is broad and extended to temperatures well above T_N in both *A* and *B* layers. Ordering of the *B* layers induced by the field orders the *A* layers as well by the interlayer interaction.

In summary, similar broadening and shift to higher temperature of the crossover with field in all layers signifies a Type I crystal. Narrow, almost unchanged crossover in *A* layers alternating with broad, shifted crossover in *B* layers signifies a Type II crystal.

3. Structure and Magnetism in κ-(BEDT-TTF)₂-Cu[N(CN)₂]Cl

 κ -ET₂-Cl is a layered spin 1/2 Heisenberg antiferromagnet with a magnetic ordering transition⁴⁾ at $T_{\rm N} = 23$ K measured⁵⁾ in H = 0. The ET molecules are arranged within layers in a 2D lattice of singly charged dimers. The electronic band is effectively half filled and the system is on the insulating side of a nearby metal–insulator Mott transition. The two chemically equivalent organic ET layers with alternating molecular orientations related by a glide plane symmetry,^{6,7)} *A* and *B* are separated by Cu[N(CN)₂]Cl polymer sheets [Fig. 2(a)]. Consequently, in the magnetic field directions $\varphi_{ab} \approx 134^{\circ}$ and 46° the ESR spectra are the same, although the contributions from *A* and *B* layers are interchanged.

The ordered state is well described by weakly ferromagnetic two-sublattice antiferromagnetic layers (Fig. 3). The isotropic exchange is two orders of magnitude larger than the



Fig. 2. (Color online) (a) Structure of κ -ET₂-Cl. Projection onto the (**a**, **b**) plane. Only two ET molecules per plane are shown for clarity. (b) Orientation of the Dzyaloshinskii–Moriya vectors \mathbf{D}_A and \mathbf{D}_B and the principal crystallographic directions. A magnetic field, **H** parallel to \mathbf{D}_A in the *A* layers is nearly perpendicular to \mathbf{D}_B in the *B* layers. (c) The ESR (derivative dispresion) spectrum at 222.4 GHz and 75 K for magnetic field, **H** || \mathbf{D}_A . The *g*-factor anisotropy splits the lines of layers *A* and *B* by 20 mT. The narrow line at higher fields is a reference.



Fig. 3. (Color online) Projection of the structure of a single layer of κ -ET₂-Cl along the long molecular axis onto the (**a**, **c**) layer plane. There is one electron on each molecular dimer. Arrows are the magnetic moments in a field **H** \perp **D** in the (**a**, **b**) plane. The weak ferromagnetism, i.e., the slight canting of $\approx 0.5^{\circ}$ along **H** in the ordered state at low temperatures is not shown. The staggered effective magnetic field, **h** = (**D**₁₂/*J*) × **H** arising from the DM interaction induces an antiferromagnetic order in the crossover temperature region above $T_{\rm N}$.

DM interaction (giving rise to the weak ferromagnetism) and more than five orders larger than other anisotropic intralayer interactions and the coupling through the polymeric sheets.⁸⁾

The measured macroscopic parameter of the in-plane isotropic exchange, $(J/2) \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j$ is $\lambda M_0 = 2J/(g\mu_B) =$ $450 \text{ T.}^{4,9)}$ The magnitude of the antisymmetric Dzyaloshinskii–Moriya (DM) exchange interaction, $(1/2) \sum_{i,j} \mathbf{D}_{ijl} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ is $DM_0 = 2D_{12}/(g\mu_B) = 3.7 \text{ T.}^{8,10)}$ [The summation is over the four first-neighbor dimers *i* and *j* (Fig. 3), l = A, B. M_0 is the T = 0 sublattice magnetization. The length of the (**a**, **b**) plane component of the DM interaction, $D_{12} = |D_{ijl}^{ab}|$ is the same for all pairs]. The DM vectors of *A* and *B* layers are approximately perpendicular: \mathbf{D}_A and \mathbf{D}_B are aligned at $\varphi_0 = 134$ and 46° respectively from **a** in the (**a**, **b**) plane.¹⁰⁾ The continuous rotational symmetry around the DM vector is nearly perfect; both the in-plane anisotropy of the exchange and the coupling between planes are very small, of the order of $1 \text{ mT.}^{8)}$

In usual antiferromagnets the order is slightly depressed by **H**. In κ -ET₂-Cl, however, the onset of the magnetic order increases significantly with H.^{11,12} Kagawa⁵ pointed out that the DM interaction is the reason for the rapid increase of the crossover temperature with magnetic field. In the presence of the DM interaction the AF magnetization remains finite above T_N in a magnetic field and there is a weak ferromagnetic magnetization along H up to high temperatures. Kagawa⁵⁾ measured by NMR the field-induced staggered static magnetic moment per sublattice site, μ_s , to temperatures well above $T_{\rm N}$. They found that a mean field model describes qualitatively the increase of the crossover temperature with H but unrealistic exchange interaction parameters are required for a quantitative agreement. As the magnetic field was along the high symmetry crystallographic axis, **a** in the experiment of Kagawa,⁵⁾ A and B layers were magnetically equivalent with the same AF magnetizations. Like the present ESR study, NMR under fields tilted in the (**a**, **b**) plane could also decide between Type I and Type II crystals.

4. Determination of the Field Induced Sublattice Magnetization

We know of no calculations of the ESR in the 2D anisotropic Heisenberg antiferromagnet. On the other hand, the excitations and the ESR spectrum of the 1D chain were extensively studied.^{3,13–16)} The DM interaction in an external magnetic field acts approximately as a staggered magnetic field, $\mathbf{h} = (\mathbf{D}_{12}/J) \times \mathbf{H}$ with an alternating sign for subsequent molecules along the chain.³⁾ (A contribution to \mathbf{h} from the in-plane alternating *g*-factor anisotropy is negligible in κ -ET₂-Cl). **H** induces an anisotropic gap in the excitation spectrum proportional to $(DH)^{1/2}$ for small *H*. The gap is largest for $\mathbf{H} \perp \mathbf{D}$ and vanishes for $\mathbf{H} \parallel \mathbf{D}$. At low temperatures **D** shifts the q = 0 ESR mode.

As shown below, the ESR in κ -ET₂-Cl resembles the ESR in 1D Heisenberg chains with a DM interaction,¹⁵⁾ the main difference is that κ -ET₂-Cl orders at a finite temperature. The similarity with the 1D chain arises from the structure. Within a single layer the joint action of the DM interaction and the magnetic field can be replaced by a staggered effective magnetic field, **h** that is uniform within lines of dimers along **a** but alternates along **c** (Fig. 3). In the mean field approximation the frequencies of the two q = 0 modes are:¹⁷⁾

$$\hbar\omega_{-} = (hJ\mu_{\rm s})^{1/2} \tag{1}$$

$$\hbar\omega_{+} = g_{0}\mu_{\rm B}H\left(1 + \frac{hJ\mu_{\rm s}}{2(g_{0}\mu_{\rm B})^{2}H^{2}}\right)$$
(2)

to the smallest order in h. Here g_0 is the *g*-factor defining the resonance frequency without the DM interaction.

The ESR frequency above T_N is given by $\omega_+ h = HD_{12}/J$ for $\mathbf{H} \perp \mathbf{D}$, and the magnetic field dependent shift of the *g*-factor is:

$$\Delta g = g_0 (DM_0/8H) \mu_{\rm s}/\mu_{\rm B}.\tag{3}$$

For $\mathbf{H} \perp \mathbf{D}$ we have h = 0 and there is no shift.

Fluctuations of the staggered magnetization at the Larmor frequency enhance the transverse spin relaxation rate, $1/T_2$

and increase the ESR linewidth, $\Delta H = (\gamma T_2)^{-1}$. In a 1D chain, the line broadening is anisotropic, roughly proportional to h^2/T^2 if $\mathbf{H} \perp \mathbf{D}$, but absent if $\mathbf{H} \parallel \mathbf{D}$. A similar anisotropic broadening of the ESR spectrum will take place in a 2D system in the crossover region above the finite T_N .

We note that there is a gap in the fluctuation spectrum below the lower frequency mode ω_{-} . While this small gap does not affect the ESR relaxation, it may have a strong effect on the NMR spin relaxation where the Larmor frequency is three orders of magnitude smaller.

5. Outline of the Experiment

We measure the magnetic order by electron spin resonance (ESR) in κ -ET₂-Cl above T_N in fields between 0.3 and 15 T. The static component of the field induced magnetization shifts the resonance while low frequency fluctations broadens the spectrum. The field induced magnetization in the paramagnetic state is very small at low field (0.3 T) except very near to T_N . μ_s is determined from the shift difference, Δg , between high field and the lowest field using Eq. (3). This novel ESR method to determine μ_s is tested by a comparison to the NMR data of Kagawa.⁵⁾

At high fields along \mathbf{D}_A (and thus perpendicular to \mathbf{D}_B) the field induced magnetic order in A and B layers is simultaneously measured as their ESR spectra are split by a small g-factor anisotropy [Fig. 2(c)]. Thus the order in Alayers that are not directly ordered by the field but are sandwiched between B layers that are directly ordered can be compared. Similarly, measurement of the ESR linewidth allows a comparison of the low frequency fluctuations of the field induced AF magnetizations of A and B layers.

These experiments show whether the order is induced independently in adjacent layers or not. We show in the following section that κ -ET₂-Cl is to a good approximation a Type II crystal where the magnetizations of a single layer depends on the direction and magnitude of **H** acting on it but not on the magnetic order in neighbouring layers. Thus, as explained in Fig. 1 the order in each layer of the crystal depends on intralayer anisotropy and not on interlayer coupling.

6. AF Magnetization in A and B Layers of κ -ET₂-Cl

6.1 ESR in κ -ET₂-Cl

Single crystals of κ -ET₂-Cl were grown by the standard electrochemical method. Crystal quality was verified by X-ray diffraction. The ESR spectrometers^{18–20)} operate at 111.2 and 222.4 GHz at BME¹⁸⁾ and at 9, 210 and 420 GHz at EPFL.^{19,20)} In discussing the ESR spectra we often refer to *H*, the approximate ESR magnetic fields instead of the fixed excitation frequency, $\omega_+/2\pi$. We denote the 9.4 GHz ESR data centered at 0.34 T as "low field" and the 111.2, 210, 222.4, and 420 GHz data centered near 4, 7.5, 8, and 15 T respectively as "high field".

The general behavior of the ESR as the crystal is cooled from the ambient temperature paramagnetic state to the fully ordered low temperature weak ferromagnetic state in a field of 8 T is demonstrated in Fig. 4. Here **H** || **c** thus *A* and *B* layers are magnetically equivalent.^{8,21,22} Well above the ordering crossover, between 250 and 40 K, the linewidth and *g*-factor are independent of magnetic field and depend little on temperature. In the crossover region, below 40 K, the ESR



Fig. 4. (Color online) Temperature dependence of κ -ET₂-Cl ESR dispersion derivative spectra at 222.4 GHz frequency and **H** || **c** magnetic field. The relative amplitudes at different temperatures are arbitrary. (a) Two antiferromagnetic resonance modes below $T_{\rm N} = 23$ K. (b) ESR in the paramagnetic region. The shift and broadening near $T_{\rm N}$ is attributed to the AF magnetization induced in the planes perpendicular to the DM vectors.

broadens and shifts to lower fields. No ESR is observed within a few degrees K of $T_{\rm N} = 23$ K. The antiferromagnetic resonance (AFMR) modes appearing at 20 K shift rapidly with lowering the temperature. The two narrow AFMR modes in the **c** direction at 4 K are shifted several T from the high temperature paramagnetic resonance. The parameters of the magnetically ordered state (in particular the in-plane anisotropy and interlayer coupling between layers) have been determined by mapping the angular dependence of the AFMR modes.⁸⁾

6.2 ESR in the crossover temperature range

The **H** and temperature (*T*) dependence of the ESR spectra were measured in detail in the crossover region above $T_{\rm N}$. The low field (9.4 GHz) ESR spectra in the **b** direction agree with the results of Yasin.²³⁾ The low field spectrum changes little with temperature above 30 K; the resonance field is independent of temperature (Fig. 6), the linewidth increases with increasing temperature up to about 40 K and is constant above (Fig. 8). As $T_{\rm N}$ is approached below 30 K, the line shifts to lower fields (higher *g*-factor) and looses intensity. The ESR could not be followed below 26 K. At 9.4 GHz the splitting of the *A* and *B* ESR lines in the (**a**, **b**) plane is not resolved.

At high fields, there is a single ESR line in the three principal directions where *A* and *B* layers are magnetically equivalent, while the *A* and *B* layer ESR lines are resolved in the (**a**, **b**) plane. To a good approximation, at field angle $\varphi_{ab} \approx 134^{\circ}$ the field in the *A* and *B* layers is oriented **H** || **D**_A and **H** || **c** × **D**_B (i.e., **H** \perp **D**_B), respectively [Fig. 2(c)]. Using the known orientation of the *g*-tensor,²⁴⁾ the lower field ESR line is assigned to the *A* layers.

The contributions of the *A* and *B* layers to the ESR spectrum in a high field along $\varphi_{ab} \approx 134^{\circ}$ are shown in Fig. 5. At high temperatures the linewidths of the *A* and *B* layers are similar, the resonance fields are temperature independent. The crossover from the high temperature paramagnetic state to the antiferromagnetic state is marked



Fig. 5. (Color online) ESR derivative dispersion spectra for **H** in the $\varphi_{ab} \approx 134^{\circ}$ direction in the crossover region at 210 GHz. Black line: experimental spectra (arbitrary amplitudes). Blue and red area are contibutions from *A* layers (with **H** || **D**_{*A*}) and *B* layers (with **H** \perp **D**_{*B*}) respectively. See text for fitting procedure. At 30 and 28 K the ESR of *B* layers is very broad and the full experimental spectrum is attributed to *A* layers. Note the shift to higher temperature of the crossover in *B* layers.



Fig. 6. (Color online) Temperature dependence of the *g*-factors (left scale) and Δg (right scale) in various fields in κ -ET₂-Cl. \blacksquare : (right scale) Δg calculated from Eq. (3) using the NMR data on μ_s at H = 7.4 T along **a** ($H_{\perp} = 5.2$ T) of Ref. 5.

by a shift to lower fields and a broadening of the line in both *A* and *B* layers but at different temperatures. The crossover in the layers depends on the orientation of the field with respect to the DM vector: in *B* layers where $\mathbf{H} \perp \mathbf{D}_B$ the crossover is at several degrees higher temperature than in *A* layers where $\mathbf{H} \parallel \mathbf{D}_A$.

6.3 Field induced static AF magnetization

We characterize in Fig. 6 the resonance fields H_{res} by the *g*-factor defined as $\hbar\omega_+ = g\mu_B H_{\text{res}}$. The field induced AF magnetization is negligible above 50 K since the *g*-factor is temperature (*T*) and **H** independent. The right scale of Fig. 6 is the difference $\Delta g = g - g_0$ where g_0 is the *g*-factor at 50 K for **H** \parallel **c** × **D**_B.

The field and temperature dependence of the g-factor in Fig. 6 shows unambiguously that the magnetization is induced independently in the A and B layers and the



Fig. 7. (Color online) Magnetic field and temperature dependence of the ESR linewidth in the crossover region measured simultaneously in the *A* and *B* layers with $\mathbf{H} \parallel \mathbf{D}_A$ and $\mathbf{H} \perp \mathbf{D}_B$ respectively.

interlayer coupling is negligible in the development of magnetic order. In *A* layers where $\mathbf{H} \parallel \mathbf{D}_A$ the AF magnetization is negligible above 30 K in all fields since the *g*-factor is independent of *H* and *T*. Below 30 K the *g*-factor increases as T_N is approached but this increase is independent of field strength. The *T* dependence of the *g*-factor in *A* layers at high fields for $\mathbf{H} \parallel \mathbf{D}_A$ is the same as at low fields for $\mathbf{H} \parallel \mathbf{b}$. In contrast, in *B* layers where $\mathbf{H} \perp \mathbf{D}_B$, Δg is anomalously field dependent: as $T \rightarrow T_N$ from above, Δg has an upturn at several degrees higher temperatures in high fields than in low field. As discussed in Sect. 4, this field dependence of the *g*-factor is explained by the field induced static staggered magnetization. The measured Δg shifts for $\mathbf{H} \perp \mathbf{D}_B$ are in excellent agreement with Eq. (3) using μ_s measured by NMR⁵ in similar fields.

6.4 Fluctuations of the field induced AF magnetization

The same conclusion is derived from the magnetic-fieldinduced ESR linewidth presented in Figs. 7 and 8. The half width at half maximum linewidths are determined from a fit to Lorentzian derivative line shapes. This is an aproximation since the spectrum is influenced by the very small but not strictly zero coupling between adjacent layers. For magnetic fields along $\varphi_{ab} = 134^\circ$, a detailed analysis of the lineshape²²⁾ reveals a small static exchange field proportional to the homogeneous magnetization below 40 K. In the present work interplane magnetic coupling was taken into account by fitting the ESR spectra to two Lorentzians with different admixtures of the absorption and dispersion components.

There are two contributions to the linewidth in the paramagnetic state below about 50 K: $\Delta H = \Delta H_0(T) + \Delta H_{\rm fl}(\mathbf{H}, T)$. $\Delta H_0(T)$ is measured at low fields. The anisotropy of $\Delta H_0(T)$ is small.²³⁾ $\Delta H_0(T)$ increases smoothly with decreasing temperature from 300 K to about 75 K and decreases below 50 K in all directions.

It is natural to assume that the **H** dependent linewidth $\Delta H_{\rm fl}(\mathbf{H}, T)$ arises from fluctuations of the staggered magnetization \mathbf{M}_s . Although in general, both longitudinal and transverse fluctuations (with respect to **H**) increase the linewidth, near $T_{\rm N}\mathbf{M}_s$ is much larger than the longitudinal magnetization. Fluctuations of the field-induced staggered



Fig. 8. (Color online) Magnetic field dependence of the ESR linewidth ΔH for **H** || **b** above T_N . The field has a large component perpendicular to the DM vector. In high field (15 T) the field induced antiferromagnetic fluctuations broaden the line well above T_N . The broadening becomes apparent at lower temperatures at 4 T. At low field (0.34 T) antiferromagnetic fluctuations are confined to a narrow region above T_N and the line broadening is not observed. Instead, the line intensity decreases rapidly close to T_N as the static antiferromagnetic phase develops.

magnetization, measured by $\Delta H_{\rm fl}(\mathbf{H}, T)$ rapidly increase at high fields as $T_{\rm N}$ is approached. At fixed temperatures near $T_{\rm N}$, the increase of $\Delta H_{\rm fl}(\mathbf{H}, T)$ with **H** is stronger than linear (Figs. 7 and 8).

Like the *g* shift, the field-induced linewidth of individual layers is independent of the magnetization in adjacent layers. There is no field-induced line broadening parallel to \mathbf{D}_A in the *A* layers where ΔH is field independent above 30 K. In contrast, in the simultaneously measured adjacent *B* layers where $\mathbf{H} \parallel \mathbf{c} \times \mathbf{D}_B$ (i.e., $\mathbf{H} \perp \mathbf{D}_B$) fluctuations broaden the line beyond observability at 30 K (Fig. 7).

Within experimental precision only the component of **H** perpendicular to **D** broadens the line. $\Delta H_{\rm fl}(\mathbf{H}, T)$ is largest in the plane perpendicular to **D**. The induced linewidth is smaller in the **b** direction than the linewidth induced by fields of the same magnitude along $\mathbf{c} \times \mathbf{D}_B$. (Along **b** only $1/\sqrt{2}$ fraction of the field is perpendicular to **D**). $\Delta H_{\rm fl}(\mathbf{H}, T)$ is isotropic in the plane perpendicular to the DM vector: it is about the same in the **c** direction where $\mathbf{H} \perp \mathbf{D}$ in all layers, and in the $\mathbf{c} \times \mathbf{D}_B$ direction where $\mathbf{H} \perp \mathbf{D}$ only in every second layer. This also supports that the magnetization is induced independently in adjacent layers.

7. Concluding Remarks

The magnetic-field-induced increase of the static and low frequency staggered magnetization, \mathbf{M}_s , in layers with $\mathbf{H} \perp \mathbf{D}$ is a 2D effect depending only on intralayer interactions of κ -ET₂-Cl. In layers with $\mathbf{H} \parallel \mathbf{D}$, both the static component of \mathbf{M}_s and its fluctuations are insensitive to the large increase of \mathbf{M}_s in adjacent layers in the crossover region above T_N . The absence of correlation between the magnetizations of adjacent layers measured in a broad range of magnetic fields suggests that interlayer correlations are unimportant in determining the phase transition temperature T_N (in H = 0); and in this sense κ -ET₂-Cl is a 2D magnet.

Finally we discuss the dimensionality of the phase transition. The absence of correlation between the magnetic

order of adjacent layers raises the question whether the phase transition at $T_{\rm N} = 23$ K in the absence of H is driven by the slightly anisotropic intralayer exchange alone, or does the interlayer coupling enforce an order in the third direction? Below $T_{\rm N}$ the weak ferromagnetism of isolated layers have a twofold degeneracy in H = 0 which is lifted even in small fields. In our experiments there is a crossover to a weak ferromagnetic phase since under magnetic fields the degeneracy is lifted even in the H || D layers by misalignment of the field, the g-factor anisotropy between layers and other small anisotropic effects. We note that although $T_{\rm N}$ was determined in H = 0 in the experiment of Ref. 5, the order in the third direction cannot be inferred from this type of experiment, either, since the ferromagnetic order was enforced at low temperatures by a magnetic field prior to the determination of T_N in H = 0. Thus the full 3D order (i.e., the order along the third direction) in H = 0 remains an open question. It may be ferromagnetic, antiferromagnetic, and this transition might even be at a temperature below $T_{\rm N}$.

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