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Kosterlitz-Thouless transition in (CH$_3$NH$_3$)$_2$.CuCl$_4$

M. Aïn

Service de Physique du Solide et de Résonance Magnétique, CEN-Saclay, 91191 Gif-sur-Yvette Cedex, France

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Résumé. Nous présentons les courbes de susceptibilité magnétique de monocristaux de (CH$_3$NH$_3$)$_2$.CuCl$_4$ faites en champs appliqués statique et basse fréquence, ainsi que des spectres de diffusion magnétique de neutrons, obtenus sur des monocristaux de (CD$_3$ND$_3$)$_2$.CuCl$_4$. De l'ensemble de ces résultats il paraît évident que de fortes fluctuations magnétiques persistent en dessous de la température de transition à $T_c = 8.77$ K et que la susceptibilité ne diverge pas avant $T = 0$ K. Ceci est en accord avec des résultats déjà obtenus [10] par diffraction de neutrons sur des monocristaux de (CD$_3$ND$_3$)$_2$.CuCl$_4$, qui ont montré qu'il y avait une mosaïque importante et que le comportement magnétique de ce cristal était de nature bi-dimensionnelle. Dans le cadre du modèle planaire, nous proposons un calcul de la susceptibilité d'un domaine bidimensionnel de taille finie, qui rend compte en dessous de $T_c$, de la variation de la susceptibilité magnétique en champ statique. Par déduction, nous identifions la transition à $T_c$ comme étant de type Kosterlitz-Thouless.

Abstract. We present magnetic susceptibility curves, made on single crystals of (CH$_3$NH$_3$)$_2$.CuCl$_4$ under static and low-frequency applied fields; also shown, magnetic neutron diffusion patterns obtained from single crystals of deuterated (CD$_3$ND$_3$)$_2$.CuCl$_4$. During the re-examination of the magnetic behaviour of this compound it became evident that strong magnetic fluctuations persist below the transition temperature at $T_c = 8.77$ K, and that the magnetic susceptibility does not diverge before $T = 0$ K. This is supported by previous results [10] obtained by neutron diffraction on single crystals of (CD$_3$ND$_3$)$_2$.CuCl$_4$, that revealed that there was a large mosaic spread and that most of the magnetic ordering in the crystal was only two-dimensional. This led us to calculate, in the framework of the planar model, the susceptibility of a finite two-dimensional domain, whose final expression reproduces, below $T_c$, the behaviour of the experimental curve in static field. Thus we deduce that the transition that takes place at $T_c$ is of Kosterlitz-Thouless type.

1. Introduction.

Methylammonium copper chloride (CH$_3$NH$_3$)$_2$.CuCl$_4$, has received much attention because of its two-dimensional magnetic character. The magnetic ion is a spin 1/2 Cu$^{2+}$ ion surrounded by 6 chlorine ions in quasi-octahedral coordination, distorted by the Jahn-Teller effect. These quasi-octahedrons are assembled in a staggered way on a plane, with their elongated axis approximately horizontal, forming a nearly quadratic array of Cu$^{2+}$ ions, figure 1. One sees that there are two kinds of Cu$^{2+}$ ions, depending upon the direction of the local 4-axis. Within the planes, Cu$^{2+}$ ions interact via a ferromagnetic exchange coupling $J$; on both sides of these planes, sticking in the alveoles formed by 4 neighbouring Cu$^{2+}$ octahedrons, we find the ammonium termination of the CH$_3$NH$_3$ groups. The crystal itself is built up by piling such electrically neutral assemblies, coupled to each other by van der Waals interactions rendering the exchange between planes of copper extremely weak.

![Fig. 1.](http://dx.doi.org/10.1051/jphys:0198700480120210300)
The compounds (CH₃NH₃)₂CuCl₄ and deuterated (CD₃ND₃)₂CuCl₄ crystallize as flat yellow platelets that cleave easily in the plane of the platelets. The colour turns to pale green at low temperature, owing to thermochromism. These crystals are very fragile, and are subject to internal stresses due to the plastic and structural deformations that occur below 300K [11, 14 bis, 33]. As a consequence, during our X-rays experiments with a precession camera on single crystals, we could not keep, at low temperature, the same orientation as at room temperature (14 bis). Let us simply recall the succession of structural transition that occurs in both compounds:

\[ T = 347 \text{ K} \quad T < 230 \text{ K} \]

Tetragonal \rightleftharpoons\text{ Orthorhombic} \rightleftharpoons\text{ Monoclinic} \quad (\beta = 92.3 \text{ at } T < 30 \text{ K})

We found for the lattice parameters: \(a = 18.65\) Å, \(b = 7.28\) Å, \(c = 7.38\) Å at room temperature; where \(a\) is the lattice parameter along the perpendicular direction to the basal planes of copper.

The following section (Sect. 2) is devoted to the description of the experimental settings where one can find an account of the calibration method used to obtain absolute measurements of the static susceptibility. Section 3 stresses the salient points of the experimental results; in section 4 a model is proposed that provides an explanation for the unusual behaviour of the static susceptibility of (CH₃NH₃)₂CuCl₄; and finally section 5 concludes this paper.

2. Experimental settings.

2.1 MAGNETIC NEUTRON DIFFRACTION. – We have performed neutron diffraction on a single crystal of (CD₃ND₃)₂CuCl₄ of approximately \(0.2 \times 4 \times 7\) mm³. The experiment was performed on a two axes spectrometer at a wavelength of \(\lambda = 2.467\) Å. A graphite filter was inserted in the incident beam to minimize the harmonics. Scans along \(h00\) were recorded at four temperatures 1.35, 4.3, 8.8 and 37.5 K, using a graphite thermometer.

2.2 MAGNETIC SUSCEPTIBILITY.

2.2.1 Static field measurements. – These were performed by using a home-made (except for the Josephson junction) SQUID, operating in a helium-4 refrigerator; we could cover the temperature range of 1.5 - 30 K using a germanium thermometer. This ensemble was shielded on the outside by a cylinder of μ-metal, another cylindrical superconducting shield was immersed in the liquid helium bath in order to screen from stray magnetic fields. The magnetic field was applied to the sample by means of a solenoid that produces \(2.5 \times 10^{-5}\) Tesla/mA. The ellipsoidal shaped sample was at the centre of the transformer coil composed of two circular loops of internal radius: \(r = 6\) mm. These two loops were connected to the signal coil which in turn was magnetically coupled to the Josephson junction of the SQUID; the transformer plus the signal coils forming together a superconducting circuit. The self-inductance of the transformer coil was lower than the one of the signal coil by at least a factor of 50, in order to minimize the field that the superconducting circuit creates on the sample, in response to any flux variation.

2.2.2 Calibration. – The magnetometer described above was calibrated as follows. In a first step, we applied a uniform static field across the transformer coil, this gave us a flux sensitivity of \(7.213 \times 10^{-11}\) Maxwell by volt on the SQUID. Afterwards we calculated the expected flux threading the coil assembly from a homogeneously magnetized ellipsoid. This procedure enabled us to obtain the magnetization (or equivalently the susceptibility) of the sample from the knowledge of the reading in volt on the SQUID.

For a paramagnetic ellipsoid having two equal minor axes of length \(2b\) and a major axis of length \(2a\), in a uniform magnetic induction \(B\), we calculated [27] that, provided that the configuration has a symmetry of revolution, the flux threading the two loops of the coil was given by:

\[
\phi = 2\pi b^2 \left[ d - 1 - \frac{1}{E^3} \left( E(Y - 1) + \frac{1}{2} (Y^2 - E^2) \right) \times \frac{\ln \frac{Y - E}{Y + E} + \frac{1}{2} (E^2 - 1) \ln \frac{1 - E}{1 + E}}{1 + d\chi} \right] B
\]

(1)

the factor 2 stands for the two loops, \(\pi b^2\) is the section of the ellipsoid in the plane of the two loops, \(d\) is the familiar demagnetizing field factor which can be expressed as:

\[
d = \frac{b^2}{2a^2E^3} \left( -2E + \ln \frac{1 + E}{1 - E} \right)
\]

(1b)

\(\chi\) is the theoretical susceptibility of this material, as will be calculated in section 4, \(E\) is the excentricity of the ellipsoid:

\[
E = \frac{1}{a} \sqrt{a^2 - b^2} \quad \text{and} \quad Y = \frac{1}{a} \sqrt{R^2 + a^2 - b^2}
\]

One can see that the measured flux \(\phi\) is proportional to the « measured susceptibility » \(\chi_m\) whose expression is [29]:

\[
\chi_m = \frac{\chi}{1 + d\chi}
\]

(2)

Provided \(\chi\) is real, the practical relationship between \(\phi\) and \(\chi_m\) reads:

\[
\phi = F \cdot \chi_m \cdot B
\]

(2b)
In these static field experiments we used an ellipsoidal sample measuring $12 \times 5 \times 4.6\ mm^3$ [25] with its major axis along the direction of easy magnetization. We assimilated this ellipsoidal sample to an ellipsoid of revolution of $12 \times 4.795 \times 4.795\ mm^3$; with this we deduced $d = 0.135$ using (1b), and we found $F = 1.981\ e^{-3}m^2$ using (1). Note however, that the calculated flux from an ellipsoid of revolution is underestimated, compared to the measured flux from the normal ellipsoid, for the reason that in the calculated geometry some magnetic moments were moved towards the axis of the loops [35]. In other words this calculation could predict a precocious divergence of the susceptibility.

By measuring the overall dimensions of the sample along several directions we estimated the departure from ideal mensurations to be of order $\pm 3\ e^{-2}\ mm$. Then from standard error calculation we obtained $\Delta dd = \pm 3.5\ %$ for the error on the demagnetizing factor.

2.2.3 Alternating field measurements. – The real $\chi'$ and complex $\chi''$ parts of the magnetic susceptibility were measured by means of a mutual inductance bridge operated at 17 Hz, with an applied magnetic induction of $1.7\ e^{-4}\ Tesla$ (1.7 Oersted). The sample used in the alternating field experiment was a single crystal, shaped into a disk of 0.2 mm in thickness and 3.4 in diameter, with its easy direction of magnetization lying in its circular section, and along the magnetic induction. This magnetometer has not been calibrated.

3. Qualitative analysis.

In what follows we want to draw attention to three points:

i) The static susceptibility does not diverge at any temperature different from 0.

ii) The complex alternating susceptibility $\chi''$ points to the fact that fluctuations persist below $T_c$.

iii) No conventional three-dimensional magnetic order takes place below $T_c$, as indicated by neutron diffraction.

These points will be emphasized in the detailed analysis of the experimental results within the three following paragraphs 3.1, 2 and 3.

3.1 MAGNETIC MEASUREMENTS ON (CH$_3$NH$_3$)$_2$. CuCl$_4$. – In this paper we often use the term susceptibility without further details, to mean the real part (when relevant) of the initial magnetic susceptibility, parallel to the easy direction of magnetisation of monocrystalline (CH$_3$NH$_3$)$_2$. CuCl$_4$. It will be clear in the text whether we refer to the measured or to the theoretical susceptibility (related by (2)). Let us proceed now with the review of the experimental results:

a) Figure 2 represents the magnetization curves of (CH$_3$NH$_3$)$_2$. CuCl$_4$ in three static fields of $10^{-5}$, $5 \times 10^{-6}$ and $10^{-6}\ Tesla$ (0.1, 0.05, 0.01 Oersted). First of all it is clear that a transition occurs at $T_c$, and that the magnetisation is rigorously proportional to the applied static field. (the vertical axis of Fig. 2 has been graduated in values of the susceptibility, and refers to the largest curve only)

![Figure 2: Magnetic susceptibility versus temperature.](image)

One remarkable feature of these curves is the negative slope of the static susceptibility below $T_c$, figure 2. Examination of relation (2) shows that whenever $\chi$ goes to infinity the « measured susceptibility » tends towards $1/d$. As a consequence, if our compound was a three-dimensional ferromagnet, below $T_c$, we ought to see a temperature independent plateau for $T < T_c$. But the fact is that our $\chi_{m}(T)$ in static field only reaches its limiting value of $1/d$ at $T = 0$, where it extrapolates to $\chi_{m}(0) = 7.246$ giving

The plot shows the magnetic susceptibility versus temperature for (CH$_3$NH$_3$)$_2$. CuCl$_4$ in three static fields: 0.1 G, 0.05 G, and 0.01 G. The vertical axis is graduated in values of 1/d, the inverse of the demagnetizing factor. The susceptibility is measured in absolute units.

The figure illustrates the static magnetization curves in three applied fields, showing a transition at $T_c$ and a rigorous proportionality to the applied field. The susceptibility is measured in absolute units, and the vertical axis is graduated in values of 1/d, the inverse of the demagnetizing factor.
\[ d = 1/\chi_{\text{a}}(0) = 0.138 \text{ which is within } 2.2\% \text{ to the calculated value of } d = 0.135. \] At \( T_c \) the susceptibility is more than 6\% beyond the calculated value. Therefore we will assume that the susceptibility only diverges at \( T = 0 \text{ K} \), but remains finite at \( T_c \) though it takes important values. As will be seen when examining the neutron scattering experiments, this hypothesis is quite consistent with the lack of ordering of the magnetization below \( T_c \). Note that Steijger et al. [10] have mentioned that their alternating susceptibility also failed to reach its maximum value of \( 1/d \) at \( T_c \), staying 5\% below the expected value.

In the early experiments with static field we measured the disk that has been used in the alternating susceptibility measurements, and we obtained the same behaviour below \( T_c \), as for the ellipsoidal sample. Furthermore, Renard has recently measured [26] the static susceptibility of a powdered sample of deuterated \((\text{CD}_3\text{ND}_3)_2\text{CuCl}_4\), using his Squid [30]; he did observed the same behaviour with a negative slope, below \( T_c = 8.8 \text{ K} \), though it was an admixture of parallel and perpendicular susceptibilities. This confirms that there is no fundamental difference between the hydrogenated and deuterated compounds.

b) In figure 3 we have the real and complex parts of the alternating susceptibility. The real part has already been discussed in (1).

We attribute the non-vanishing complex susceptibility below \( T_c \), to the existence of fluctuations in the system of spins; recall that the fluctuation-dissipation theorem relates the space and time fourier transforms of the complex susceptibility \( \chi' \) to the correlations between magnetic moments, according to the relation:

\[
\langle m^2(f) \rangle = 2k_B T V \frac{\chi''(f)}{f} \tag{3}
\]

where \( m(f) \) is the fourier transform of the magnetic moment, and \( V \) the volume of the sample.

c) \( \text{Cu}^{2+} \) ion in distorted octahedral fields is known to have a slight anisotropy [15], due to incomplete quenching of the orbital moment. The crystalline anisotropy appears through differences in the values of \( g_\perp \) and \( g_\parallel \); reaching 20\%. Wong et al. [22] has found by EPR : \( g_\parallel = 2.27 \) and \( g_\perp = 2.05 \) in \((\text{CH}_3\text{NH}_3)_2\text{CuCl}_4\), \( g_\parallel \) being the value along the dashed-dot line of figure 1a.

In order to estimate the anisotropies of this system we performed two spin-flip experiments, the first one with an applied field perpendicular to the basal planes of figure 1, and the other one with an applied field lying in these planes, perpendicularly to the direction of easy magnetization. We found respectively \( H_{\text{out}} = 0.1 \text{ Tesla for the anisotropy that forces the spins into the basal planes, and } H_{\text{in}} = 5 \times 10^{-3} \text{ Tesla for the anisotropy that tends to align the spins along the easy axis in the planes. This low in-plane anisotropy is an overall result that is originated by the staggered structure shown in figure 1, nevertheless it is quite effective with respect to the exchange coupling } J \text{ of order 30 Tesla, that tends to align the spins parallel to one another. Hence, it is expected that } (\text{CH}_3\text{NH}_3)_2\text{CuCl}_4 \text{ behaves like a soft magnetic material.}

3.2 NEUTRON DIFFRACTION PATTERNS. – We have performed scans along direction \((h00)\) in reciprocal space, perpendicularly to the basal planes of figure 1 where the spins are lying. On figure 4 the data are presented as difference patterns for scans from \( h = 0.5 \) to \( h = 3.7 \), obtained above and below the transition temperature, along \((h00)\) direction in reciprocal space. Therefore, the reflections are due solely to magnetic scattering ; and the negative background, represents the decrease in paramagnetic scattering upon crossing the transition temperature.

On the patterns of figure 4 one can see a large
Fig. 4.— Neutron diffraction data from (CD₃ND₃)₂CuCl₄. Temperature difference patterns for scans along (h00), from h = 0.5 to h = 3.7.

diffuse scattering that persists below the transition temperature, surmounted by a little (200) magnetic bragg peak. It appeared impossible to extract any magnetic contribution from the (400) bragg peak; furthermore, the magnetic (200) bragg peak is so feeble, that it actually represents only 2% of the (400) nuclear bragg peak; comparison with (200) nuclear bragg peak is therefore not relevant because it is accidentally nearly extinguished in (CD₃ND₃)₂CuCl₄ though being quite intense in the hydrogenated compound. As a reference we could mention that the ferromagnetic as well as the nuclear contribution to the (200) peak were totally invisible on the diffraction pattern from a fairly important amount of powdered (CD₃ND₃)₂CuCl₄; although the antiferromagnetic peak of CuO for instance, is quite visible at higher angles, on the same spectrometer, with nearly the same amount of powder.

The diffuse magnetic scattering along (h00) direction in reciprocal space goes smoothly to zero above h = 2; it reveals that the ordering is mainly two-dimensional in the (200) planes, with short range coupling between these planes; the little Bragg peak being due to a slight alignment of the magnetic moments induced by the crystalline anisotropy. These observations are quite consistent with the existence of a complex susceptibility. Finally, scans along directions perpendicular to (h00) at point (200) in reciprocal space, revealed the absence of magnetic diffusion, outside direction (h00).

It is worthwhile to compare our scans of figure 4, to Hirakawa’s scan, obtained on K₂CuF₄ (another magnetic quasi two-dimensional copper compound) shown in figure 4 of reference [34]. On the latter, one can see well developed peaks, at the transition temperature, announcing three dimensional order, in crude contrast with our observations on (CD₃ND₃)₂CuCl₄ reported in figure 4. Steijger et al. [10], has also reported that the (200) (or 002) if we use their convention) magnetic peak on (CD₃ND₃)₂CuCl₄ was within 5-10% of what they expected.

3.3 COMMENTS. — Ain et al. [1], Renard et al. [2], and de Jongh et al. [3-11], have measured the susceptibility of (CH₃NH₃)₂CuCl₄ in alternating field. They proposed values for γ the critical exponents of the susceptibility and they localized cross-over regions. References [2] and [9] have proposed respectively γ = 1.23 and γ = 1.25 which is the value predicted for the three-dimensional Ising model near Tc. All these authors admitted that the susceptibility of this compound was diverging at Tc.

To conclude this section devoted to the description of the magnetic features of (CH₃NH₃)₂CuCl₄ and (CD₃ND₃)₂CuCl₄ we want to emphasize that we now have serious doubts about these systems being three-dimensional ferromagnets. We believe that they are more likely planar two-dimensional ferromagnets, relying on the negative slope of the susceptibility and the look of the magnetic Bragg peak. The reason why susceptibility does not diverge is attributable to the fact that correlations are interrupted by cracks in the (200) basal planes. These cracks occur below 230 K when the structure goes from orthorhombic to monoclinic, because we then have two ways to lean the a axis, but both are nucleated in different points of the (200) basal planes. This phenomenon is responsible for mosaic in the single crystals (14 bis, 11).

4. Quantitative analysis.

In the preceding paragraph we explained the mechanism that divides the basal planes of the magnetic ions into small domains. In this section we aim to connect this feature with the negative slope of the static susceptibility between 0 K and Tc. To achieve this we calculate the susceptibility of a circular domain, covered by a square array of magnetic moments, assuming a decoupling between domains in the same basal plane as well as in neighbouring planes (as suggested by neutron scattering experiments). The magnetic moments are ferromagnetically coupled by an exchange interac-
tion \( J \), they lay in the plane of the domain under effect of the out of plane anisotropy, the in-plane anisotropy will be neglected. Temperature will be supposed sufficiently low, and spins \( s \) will be considered as classical vectors.

We are now meeting with the conditions of the planar model that has been thoroughly examined by Kosterlitz and Thouless [16,17], Villain [18], Jose et al. [28]. Briefly we can say that this model displays a phase transition at \( T_c \), where pairs of vortex-antivortex unbind in the high temperature phase to produce free vortices. Villain [18] has rigorously shown that the total Hamiltonian of this system separates into two parts:

- \( H_h \) a harmonic Hamiltonian that does not display any phase transition, but accounts for the properties of the system up to 0.9 \( T_c \).
- \( H_v \), the second part, appears during manipulations on the total Hamiltonian, it describes the system of vortices and displays the phase transition at \( T_c \).

Berezinsky [19] and Sarma [20] have calculated the two-spin correlation function for this model, within the harmonic approximation. They obtained:

\[
\langle s(R_1)s(R_2) \rangle = s^2 \langle \cos \theta(R_1) \rangle \cos \theta(R_2)
\]

\[
\cos \theta(R) = \frac{r}{b} \eta
\]

\[
\langle s(R_1)s(R_2) \rangle = \frac{s^2}{2} \left( \frac{r}{b} \right)^{-\eta}
\]

\( \theta(R) \) is the spin deviation from a reference axis, \( r \) is the separation between the two spins at \( R_1 \) and \( R_2 \), and \( \eta = k_BT/4\pi JS^2 \). Result (4) still holds when the two-dimensional domain is finite as is the case here. Intuitively one can say that the only modes of magnons that one has to take into account in this calculation are those whose wavelength is shorter than \( r \), this itself warrants their existence. As to the vortices, they are very scarce below \( T_c \) and bound in pairs of opposite sign [16,17]. At large distances, no disturbance is brought about by bound pairs of vortex-antivortex, since their contribution to spin deviation is vanishingly small. This image illustrates why the harmonic Hamiltonian \( H_h \) alone is sufficient for describing the magnetic system in the temperature range below \( T_c \).

With expression (4) for the correlation function, we show in the appendix how to calculate the susceptibility per ion in a circular two-dimensional domain. This gives for the susceptibility per unit volume:

\[
\chi = \beta N g^2 \mu_B^2 \left( \frac{2}{4-\eta} \right) \Gamma \left( \frac{3-\eta}{2} \right) \frac{1}{\Gamma \left( \frac{4-\eta}{2} \right)} n^{-\eta}
\]

Where \( N \) is the number of magnetic moments per unit volume. It should be mentioned that the spuriuous divergences that appear in (5) for \( \eta = 2 \) and 4 are far beyond the temperature range of validity of this calculation they bear no physical meaning. Actually \( \eta < 1.4 \times 10^{-2} \) in the region of interest between 0 and 10 K.

In view of numerical applications we will take \( N = 4.08 \times 10^{-27} \) at/m³ (i.e. : 4 Cu²⁺ ions \( S = 1/2 \) in each elementary cell of volume \( 18.6 \times 7.1 \times 7.4 \times 10^{-30} \) m³ at 4.2 K). Anticipating on our conclusions we identify the transition at \( T_c \) as the one that was discovered by Kosterlitz and Thouless. In our numerical calculation we shall use a result obtained by Villain [18]: \( k_B T_c = 1.7 JS^2 \), which gives \( J = 20.6 \) K. This value is in good agreement with experimental result from : (i) N.M.R \( J = 16.5 K \) (23), \( J = 18.5 K \) [24] ; (ii) high temperature expansions of the susceptibility \( J = 19.2 K \) [6].

Carrying (5) in (2) gives us an analytic expression for the measured susceptibility. Figure 5 represents \( \chi_m \) versus \( T \), for different values of \( n \). One can see that the curve with \( n = 400 \) reproduces very well the experimental points below \( T_c \). The distance between 2 magnetic moments being nearly 5 Å we obtain an estimate of 0.2 microns for the mean size of these two-dimensional domains, which is quite realistic.

\[
\chi_m(\ln T)
\]

Fig. 5.— The solid curves are the calculated static susceptibility \( \chi_m \) versus temperature, for different values of the size \( n \) of the circular domain. The open circles are the measured susceptibility in a static field of \( 10^{-5} \) Tesla (0.1 Oersted).

5. Conclusion.

Owing to the excellent agreement of our model with the experimental curve between 0 K and \( T_c \), we conclude that the harmonic Hamiltonian \( H_h \) of the planar model is well-suited to describe the magnetic system in hand. Furthermore we infer that the
transition taking place at $T_C$ is of the type predicted by Kosterlitz and Thouless and described by the second part $H_2$ of the total Hamiltonian of the planar model. The vortex-antivortex pairs dissociate at $T_C$ to form a gas of free vortices, as a consequence the two-spins correlation function drops abruptly, and so does the susceptibility. The reason for this is that each free vortex introduces now a rotation of roughly $\pi$ between pairs of spins, of which separation vector $r$, passes near its core. Note in passing that the identification of the Kosterlitz-Thouless transition has been done with the complicity of the cracks in the (200) basal planes. It would have been more difficult to gain insight on what was going on if the susceptibility were more diverging due to two-dimensional domains of greater extent or to the burst of three-dimensional order.

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Appendix A.

We show, in this appendix, the derivation of the susceptibility of a circular domain of radius $R$, assuming that the magnetic moments are on a quadratic array of parameter $b$, and that the correlation function, between two magnetic moments decreases as a power law of $1/r$:

$$\langle m_1, m_2 \rangle = g^2 \mu_B^2 s^2 \langle \cos \theta(R_1), \cos \theta(R_2) \rangle = \frac{[g^2 \mu_B^2 s^2]}{(r/b)^{-\eta}} \quad (A.1)$$

Where $m$ is the magnetic moment and $m = g \mu_B s$; $R_1, R_2, \theta_1, \theta_2$ are the polar coordinates of the moments $m_1$ and $m_2$, and $r = R_2 - R_1$. The total susceptibility $\chi_t$ of this domain is:

$$\chi_t = \beta g^2 \mu_B^2 s^2 \sum_{ij} \langle \cos \theta(R_i), \cos \theta(R_j) \rangle \quad (A.2)$$

We can replace the sums by integrals in the usual way $\Sigma \rightarrow 1/b^2$ thus obtaining for $\chi_t$:

$$\chi_t = \beta g^2 \mu_B^2 s^2 \frac{1}{2} \int_0^{2\pi} \int_0^R \left| r^{-\eta} R_1 dR_1 d\theta_1 R_2 dR_2 d\theta_2 \right. \right. \quad (A.3)$$

where the summations run over the surface of the circle $C$. We then replace the variables $R_2$ and $\theta_2$ by $r$ that has already been defined and $\theta$ which is the angle between $R_1$ and $r$. With the help of ordinary relations in the triangle $(r, R_1, R_2)$ we calculate that the Jacobian of this variable change is $J = r/R_2$. We are now left with:

$$\chi_t = \beta \frac{g^2 \mu_B^2 s^2}{2 b^{4-\eta}} \int_0^{2\pi} \int_0^R \left( 1 - \frac{r}{2 R} \right)^{\eta - 1} \frac{r^{1-\eta}}{2} \sqrt{4 R^2 - r^2} \, dr \, d\theta \quad (A.4)$$

Let us start with the integration with respect to $R_1$ and $\theta_1$. This integration runs over the area of intersection of two circles $C$, and separated by $r$. Writing this down, we still have to integrate over $r$ and $\theta$:

$$\chi_t = \beta \frac{g^2 \mu_B^2 s^2}{2 b^{4-\eta}} \int_0^{2\pi} \int_0^R \left( 1 - \frac{r}{2 R} \right)^{\eta - 1} \frac{r^{1-\eta} \sqrt{4 R^2 - r^2}}{2} \, dr \, d\theta \quad (A.5)$$

These integrals are tabulated. We found for the first one:

$$I_1 = \frac{2}{2 - \eta} \frac{\pi R^2}{(2 R)^{2-\eta}} \frac{\Gamma\left( \frac{3 - \eta}{2} \right)}{\Gamma\left( \frac{4 - \eta}{2} \right)} \quad (A.6)$$

and for the second:

$$I_2 = \frac{\pi}{2} \frac{R^4}{(2 R)^{4-\eta}} \frac{\Gamma\left( \frac{3 - \eta}{2} \right)}{\Gamma\left( \frac{6 - \eta}{2} \right)} \quad (A.7)$$

Where $\Gamma(\cdot)$ is the Gamma function. Summing $I_1$ and $I_2$, and dividing by $\pi R/b^2$ the number of magnetic moments in the circular domain, we obtain the susceptibility per ion:

$$\chi_t = \beta \frac{g^2 \mu_B^2 s^2}{2 b^{4-\eta}} \left( \frac{2 \sqrt{\pi}}{(4 - \eta)(2 - \eta)} \right) \frac{\Gamma\left( \frac{3 - \eta}{2} \right)}{\Gamma\left( \frac{4 - \eta}{2} \right)} \quad (A.8)$$

$n = 2 R/b$ being the number of interatomic spacings along the diameter of the circular domain. On going from (A.7) to (A.8) we made use of two results on Gamma functions:

$$\Gamma(1/2) = \sqrt{\pi} \quad \text{and} \quad \Gamma(p + 1) = p \Gamma(p) \quad (A.9)$$
References

[25] The sample was given by Pr J.P. Renard, Université Paris XI, Bât. 220, 91405 Orsay, France.
[26] Private communication.
[27] To be published.
[29] In c.g.s u.e.m units one ought to replace $\chi$ by $4\pi \chi$.
[35] Consider a coil assembly with a small sample on its axis at a distance $R$ away. The flux from the sample which threads the coil may be shown from reciprocity [36] to be:

$$\Phi = \int_{\text{sample}} \mathbf{m}(r) \cdot \mathbf{h}(r + R) \, d^3r$$

where $\mathbf{m}(r)$ is the magnetization density at the point $r$ from the centre of the sample and $\mathbf{h}(R+r)$ is the field which would be produced at $r$ if unit current flowed in the coil. In our experiment $\mathbf{m}(r)$ is parallel to the $z$ axis and $h_z(R+r)$ has a minimum at $r = 0$.