Free Vortices in the Quasi-Two Dimensional XY Antiferromagnet $BaNi_2(PO_4)_2$?

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Non-linear excitations are expected to play an important role in low dimensional magnetic systems. The existence of soliton excitations in magnetic chains is now well established theoretically and experimentally. In two dimensions non-linear excitations have also been predicted to be important, at least for the so-called XY model where the spins are mainly confined within the magnetic plane. As shown by Kosterlitz and Thouless (KT) and Berezinskii [1], a topological ordering, associated with the pairing of vortex excitations, is expected to occur at a finite temperarure T_{KT}. Above T_{KT}, the unbinding of the vortex pairs gives rise to a gas of freely moving vortices. Recently, it was shown that the vortex motion results in a flipping process of the ordered spins [2]. In the fluctuation spectrum, this flipping process yields a characteristic central peak —the flipping mode. As shown for the case of solitons in antiferromagnetic (AF) chains where a similar effect was observed, the nuclear magnetic resonance (NMR) technique is well suited to probe such a narrow peak, centered at $\omega \approx 0$ [3].

In real systems, the KT transition and the topological order can never be observed. This is due to the coupling between planes which, always, induces a three-dimensional (3D) order at a temperature $T_N > T_{KT}$. However, for a very small interplane coupling, T_N and T_{KT} can be very close and vortex excitations, if they do exist, can only be observed above T_N. The application of an external magnetic field H may change the properties and the dynamics of the vortices. This latter point has been discussed recently for the case of ferromagnets [4]. For planar antiferromagnets, one expects the effect of a field H to be small, if it is applied perpendicular to the magnetic plane ($H_{\perp}$). In that case, at least for moderate field values ($H \ll J$) the effect of a field is essentially to reinforce the planar character of the spin system.

In the present work, we report on nuclear spin-lattice relaxation time (T_1) measurements performed on the compound $BaNi_2(PO_4)_2$ [5]. As shown on fig. 1, a spectacular diverging

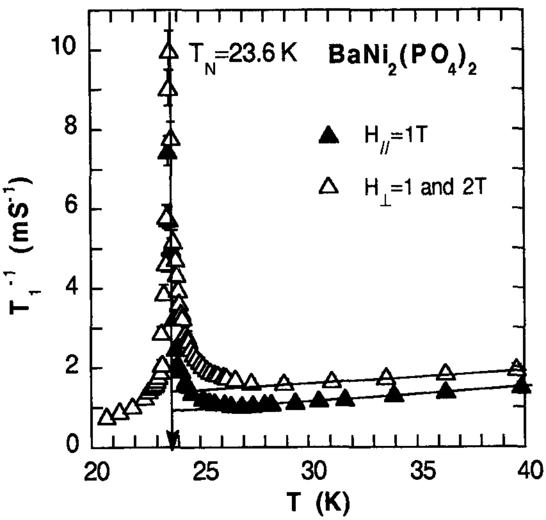

Fig.1 : Nuclear spin-lattice relaxation rate $1/T_1$ of ^{31}P in $BaNi_2(PO_4)_2$ for H perpendicular and parallel to the XY magnetic plane.

behaviour is observed for $1/T_1$ as the 3D transition (T_N≈23.6K) is approached. The data for $T>T_N$ are tentatively analysed within a model of freely moving vortices. The extrapolation of this model to the case where the field is parallel to the plane ($H_{//}$) is also discussed.

The compound $BaNi_2(PO_4)_2$ is a good quasi-2D planar antiferromagnet [6]. Its magnetic structure consists of 2D layers of Ni ions (with spin S=1), located on a honeycomb lattice of parameter a=2.8 Å. The in-plane exchange coupling is antiferromagnetic with the (average) value J≈11K. The planar character observed at low temperature results from a large single ion anisotropy D≈7.3K. The 3D magnetic ordering occuring at T_N≈23.6K is attributed to a very small interplane coupling J' ($J'/J \approx 10^{-3} — 10^{-4}$). As shown by Regnault et al. [6], just above T_N ($T-T_N$<10K), the short range order is mainly two dimensional and can be analysed in the model of the KT transition. In that case, it was deduced that T_N should be very close to T_{KT} : $T_{KT} \approx 0.95—0.98\ T_N$ [6].

Our T_1 measurements have been performed on the ^{31}P ions, as a function of the temperature and for different values and orientations of the external field. In all cases, the same kind of divergence for $1/T_1$ is observed as $T \to T_N$. However, while the data are the same for $H_\perp$=1T and 2T, they appear to be strongly field dependent for $H_{//}$. Since we want to focus on the diverging part of $1/T_1$, we consider only the contributions above the lines drawn on fig. 1. The data to be

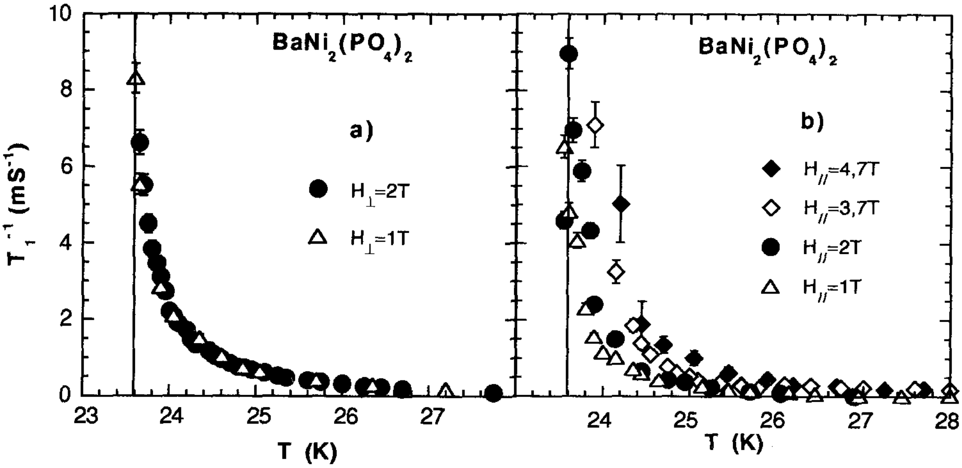

Fig.2 : Contributions from the expected critical fluctuations to $1/T_1$ for H perpendicular (a) and H parallel (b) to the XY plane.

analysed are shown in fig. 2. These values are expected to describe mainly the critical fluctuations.

For the phosphorus nuclear spins, the relaxation rate $1/T_1$ is given by $1/T_1=\Sigma_{\alpha q}A_q^{\alpha}S^{\alpha}(q,\omega_N)$ where the A_q^{α} are geometrical coefficients associated with the hyperfine coupling and the $S^{\alpha}(q,\omega)$ are the dynamical structure factors which describe the magnetic fluctuations. ω_N is the nuclear Larmor frequency (ω_N<80MHz) and q is a wavevector of the Brillouin zone. For such a 2D system, α=xy and α=z refer to the in-plane and out-of-plane fluctuations, respectively. According to the model of freely moving vortices proposed by Mertens et al. [2], one expects the main contribution to $1/T_1$ to be given by the flipping mode of the in-plane fluctuations. In reciprocal space, this flipping mode is limited to a narrow domain of the order of $\Delta q \approx \xi^{-1}$ where ξ is the 2D correlation length. Over this domain, A_q^{xy} is practically q independent ($A_q^{xy} \approx A^{xy}$) and $1/T_1$ can be written: $1/T_1 \approx A^{xy}\Sigma_q S^{xy}(q,\omega_N) = A^{xy}S^{xy}(\omega_N)$ where $S^{xy}(\omega)$ is the spectrum of local fluctuations. It has a simple Lorentzian shape with a characteristic width γ which measures the rate of the spin flippings induced by the moving vortices. Finally :

$$1/T_1=A^{xy}.\gamma \;/\; (\gamma^2+\omega_N^2) \approx A^{xy}/\gamma \qquad (1)$$

for $\omega_N \ll \gamma$. The flipping rate is evaluated to be :

$$\gamma = \sqrt{\pi n_v}.U \qquad (2)$$

where

$$n_v = 1/(2\xi)^2 \qquad (3)$$

is the density of the free vortices [7]

$$n_v = (2\xi_0)^{-2} \exp(-2b/\sqrt{\tau})$$

and U the vortex velocity [8]:

$$U = \sqrt{(\pi/2)(JS^2a^2/h)^2 n_v \, Ln\{k_B T_{KT}/(JS^2 n_v a^2)\}} \qquad (4)$$

In these expressions $\tau = T/T_{KT}-1$ and $\xi_0 \approx$ a. The parameter b is not known accurately. The KT theory predicts $b = \pi/2 \approx 1.57$ [1] ; however recent numerical simulations yield much smaller values: b≈0.5—0.3 [4,9]. The data of figure 2 are analysed with Eq.1, where A^{xy}, b and T_{KT}/T_N are considered as adjustable parameters. Fig.3 gives a few exemples of resulting fits yielding

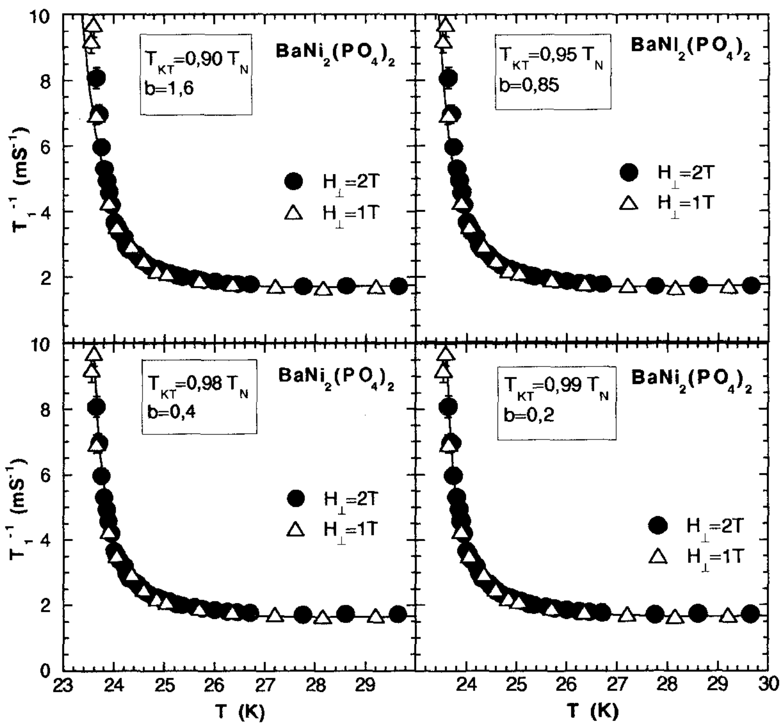

Fig.3 : Comparisons between theory (Eq. 1) and experimental data for different values of A^{xy}, b and T_{KT}/T_N

T_{KT} (K)	T_{KT}/T_N	b	A^{xy} (rad^2S^{-2})	γ (GHz) pour T=24 K
21,2	0,90	1,6	6 10^{11}	0,027
22,4	0,95	0,85	5 10^{12}	0,23
23,1	0,98	0,4	7 10^{13}	3,22
23,3	0,99	0,2	3 10^{14}	13

Table I : Values of the fitting parameters A^{xy}, b and T_{KT}/T_N corresponding to the different fits of fig. 3.

the values given in table I. The values for the flipping rate γ at 24 K are also reported in table I. A comparison with a recent neutron investigation [10] ($\gamma \approx 10$ GHz at 24 K) allows us to conclude that the values to be retained for T_{KT}/T_N, b and A^{xy} range certainly in between the values reported in the two last lines of table I. For the geometrical coefficient A^{xy} the corresponding values agree also with the evaluation we have made from the frequency shift of the NMR line : $A^{xy} \approx 10^{14}$ rad^2S^{-2}.

For $H_{//}$, a divergence is also observed for $1/T_1$, which is seen to be field dependent (fig. 2b). At low field ($H_{//} \approx 1T$), the vortex model defined above remains valid with similar values for T_{KT}/T_N and b. For larger field values, the observed increase of the divergence can be interpreted as essentially due to a slowing down of the flipping process: γ is reduced by the field $H_{//}$ (see Eq.2). As shown on fig.4, the 3D ordering temperature [10] is observed to increase slightly with $H_{//}$: for $H_{//}$ = 6 T, $\Delta T_N/T_N \approx 1.6\%$. Since T_N is proportional to the square of the 2D correlation lentgh ξ this effect corresponds to an increase of ξ. In the vortex model (see Eq. 3), this means that the vortex density n_V is decreasing with $H_{//}$: for $H_{//}$=6 T, the relative decrease of n_V is very small ($\Delta n_V/n_V$=-1.6%). Therefore, if the vortex density does not change very much, it is the vortex velocity which is reduced by the field. From the $1/T_1$ data, we can deduce the field dependence of U. The corresponding values are shown on fig.5, where they are compared to

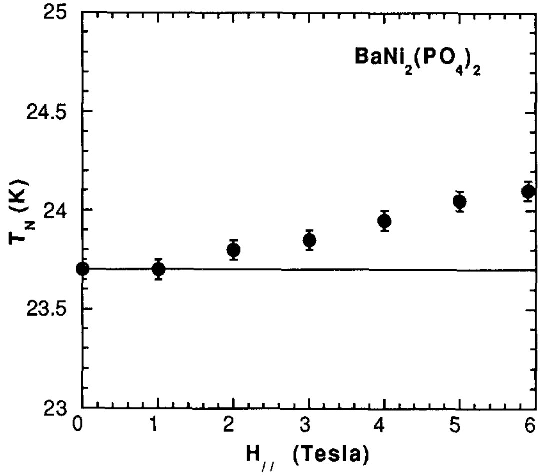

Fig. 4 : The 3D ordering temperature T_N as a function of $H_{//}$, observed by neutron scattering measurements [10].

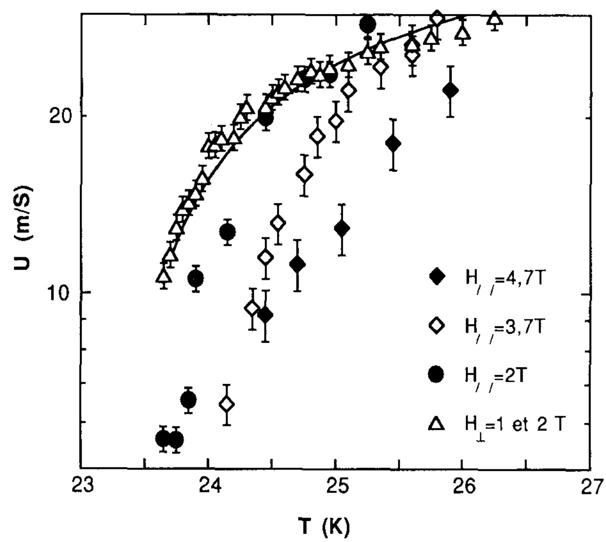

Fig. 5 : Vortex velocity U deduced from the $1/T_1$ data of fig.2. The full line represents the theoretical expression given by Huber.

the velocity given by Eq.4. In antiferromagnets, a field $H_{//}$ appears to reduce appreciably the vortex velocity. This conclusion differs from that of the ferromagnetic case [4].

The divergence of $1/T_1$ observed in $BaNi_2(PO_4)_2$ when approaching TN is well explained by a model of free moving vortices. The values obtained for b (b=0.4—0.2) is in reasonable agreement with the recent numerical simulations of Mertens et al. [4;9]. For H=0, the vortex velocity is in accord with Eq.4 given by Huber [8]. However, it is observed to be strongly field dependent.

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