# MAGNETIC PROPERTIES OF THE LAYERED NICKEL COMPOUNDS $BaNi_2(PO_4)_2$ AND $BaNi_2(AsO_4)_2$

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Neutron diffraction and specific heat measurements on the layered compounds $BaNi_2(PO_4)_2$ and $BaNi_2(AsO_4)_2$ are reported. They order antiferromagnetically below 24.05 and 18.65 K, respectively, with collinear structures defined by the wave vector $\boldsymbol{k} = 0$ and $\boldsymbol{k} = [\frac{1}{2}0\frac{1}{2}]$. The critical behaviour and the low temperature properties have been studied.

The layered compounds $BaNi_2(PO_4)_2$ and $BaNi_2(AsO_4)_2$ crystallize with a rhombohedral unit cell ($R\bar{3}$), the lattice parameters are $a = 4.82$ and 4.95 Å, $c = 23.38$ and 23.60 Å, respectively [1, 2]. They are isomorphous of the cobalt compounds in which the bidimensional character of the magnetic interactions has been evidenced [3–5]. The crystallographic structure consists of (001) magnetic planes in which $Ni^{2+}$ ions are located in a honeycomb lattice. The present paper gives a calorimetric and a neutron scattering study in order to investigate the magnetic properties of these nickel compounds.

Specific heat experiments have been performed using sintered powder samples. The nonmagnetic compounds $BaMg_2(PO_4)_2$ and $BaMg_2(AsO_4)_2$ have been measured in order to determine the lattice contribution. Elastic and quasi-elastic neutron experiments have been carried out at the Siloe reactor using a two-axis spectrometer with a wave length of 2.4 Å. The crystals were grown using a flux method and were oriented with the $b$-axis vertical.

For both compounds the magnetic contribution to the specific heat is reported in fig. 1. Sharp lambda type anomalies are observed at $T_N = 24.0$ K for $BaNi_2(PO_4)_2$ and $T_N = 19.0$ K for $BaNi_2(AsO_4)_2$, indicating a three dimensional ordering. Above this transition, the important magnetic contribution shows that strong correlations persist. At low temperatures the specific heat can be fitted by a $T^2$ law for both compounds (fig. 2), but the thermal variation is much larger for the arseniate than for the phosphate compound.

Elastic neutron scattering experiments have established that both compounds are collinear antiferromagnets; the magnetic structures are drawn in fig. 1. They correspond to a wave vector $\boldsymbol{k} = [000]$ for $BaNi_2(PO_4)_2$ and $\boldsymbol{k} = [\frac{1}{2}0\frac{1}{2}]$ for $BaNi_2(AsO_4)_2$. In the two compounds the phase angle between the two Bravais lattices of the honeycomb lattice is equal to $\pi$ and the magnetic moments are located in the magnetic plane ($X-Y$ system). The value of the magnetic moments is found to be $(2.0 \pm 0.2)\mu_B$ at $T = 4.2$ K, as expected for a spin $S = 1$.

From the thermal variation of the magnetic peak intensities, given in fig. 3, we deduce an ordering temperature at $T_N = (24.05 \pm 0.1)$ K for $BaNi_2(PO_4)_2$ and $T_N = (18.65 \pm 0.05)$ K for $BaNi_2(AsO_4)_2$ in agreement with specific heat results. Close to $T_N$ the intensities have been fitted to a power law in order to determine the critical exponent $\beta$. We get $\beta = 0.230 \pm 0.01$ for $BaNi_2(PO_4)_2$ and $\beta = 0.135 \pm 0.01$ for $BaNi_2(AsO_4)_2$; these values are much smaller than the value $\beta = 0.333$ associated to a three dimensional $X-Y$ system.

Quasi elastic neutron scattering experiments

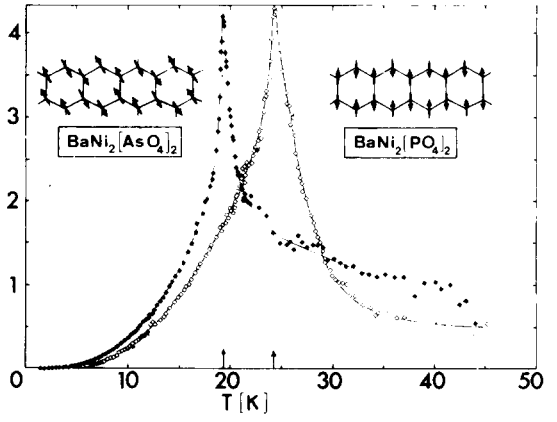


Fig. 1. Magnetic specific heat of $BaNi_2(PO_4)_2$ and $BaNi_2(AsO_4)_2$.

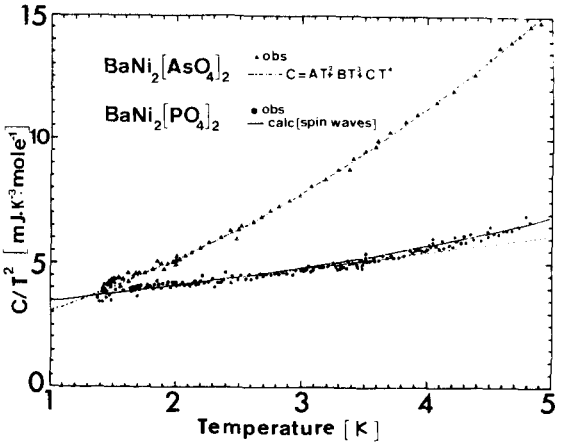


Fig. 2. $C/T^2$ vs. $T$ for $BaNi_2(PO_4)_2$ and $BaNi_2(AsO_4)_2$.

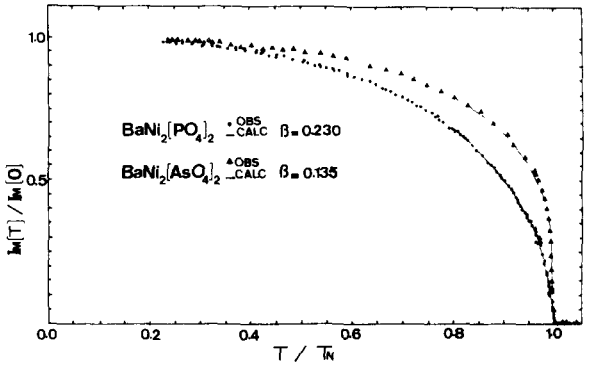


Fig. 3. Thermal variation of the intensity of a magnetic superlattice peak in reduced units for $BaNi_2(PO_4)_2$ and $BaNi_2(AsO_4)_2$.

have evidenced two-dimensional correlations up to about $2T_N$. Close to $T_N$ the correlation length follows a power law with a critical exponent $\nu = 1.5 \pm 0.1$ for $BaNi_2(PO_4)_2$ and $\nu = 1.0 \pm 0.1$ for $BaNi_2(AsO_4)_2$.

The $T^2$ contribution to the magnetic specific heat can be accounted by spin wave excitations propagating in the magnetic planes. For $BaNi_2(PO_4)_2$ the theory predicts two spin wave dispersion curves. For small $q$-values, one is linear ($\omega(q) \propto q$) and the other one has a gap ($\omega(q) \propto \Delta + aq^2$). These results have been confirmed by inelastic neutron scattering experiments [6]. Such a spectrum gives rise to a specific heat of the form:

$$C_m = AT^2 + Be^{-\Delta/T}(2\Delta + 2T + \Delta^2/T)$$

which fits the experimental data very accurately ($\Delta \approx 30$ K). The measured spin wave spectrum can be well accounted by considering magnetic interactions of Heisenberg-type with exchange integrals between first, second and third neighbors equal to $J_1 = -18$ K, $J_2/J_1 = 0.40$ and $J_3/J_1 = 0.55$ and a one ion anisotropy of $X-Y$ type ($DS_z^2$) with $D = 1.8$ K [6]. At $T = 0$ K, the stability of the magnetic structure of $BaNi_2(PO_4)_2$ is well explained with these values.

The compound $BaNi_2(AsO_4)_2$ is more complex to understand. Recent inelastic neutron scattering experiments [6] indicate that a gap at $q = 0$ exists also for the acoustic magnons. This gap can be explained by a dipolar anisotropy in the plane; thus the magnetic behaviour would be rather of Ising type than of $X-Y$ type. This result is also supported by the experimental values of the critical exponents ($\beta = 0.135$, $\nu = 1.0$) which are very close to those of the 2D. Ising system ($\beta = 0.125$, $\nu = 1.0$). $BaNi_2(PO_4)_2$, however, seems to be a good example for the $X-Y$ system with a $d = 2 + \epsilon$ dimension if we analyse the value of the critical exponents. The scaling laws give a relation between the ratio $2\beta/\nu$ and the space dimensionality: $2\beta/\nu = \epsilon + \eta$.

For an $X-Y$ system in $2 + \epsilon$ dimension calculation, using the renormalisation group theory [7], give a relation between $\nu$ and $\epsilon$: $\nu = 1/2\sqrt{\epsilon}$.

For $BaNi_2(PO_4)_2$ ($\beta = 0.230$, $\nu = 1.5$) we get $\epsilon \approx 0.1$. Thus, if we put the value $\eta = 0.25$ obtained for a 2D $X-Y$ system [8] in the scaling law we get a value $2\beta/\nu = 0.35$ which is very close to the observed value $2\beta/\nu = 0.31$. So $BaNi_2(PO_4)_2$ as $BaNi_2(AsO_4)_2$ has a 2D critical behaviour and the large value of $\nu$ is consistent with a $X-Y$ system in 2.1 dimension.

## References

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