THE EFFECT OF MAGNETIC AND NON-MAGNETIC DILUTION ON THE MAGNETIC ORDERING IN THE HEXAGONAL ANTIFERROMAGNET CsMnBr$_3$

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Abstract. - Elastic neutron scattering measurements on the hexagonal antiferromagnet CsMnBr₃ showed that $T_N$ falls steeply with concentration $x$ of the diamagnetic impurities ($Mg^{2+}$), becoming zero at $x = 0.14$; the critical exponent of the magnetisation, $\beta$, increased from 0.19(1) ($x = 0$) to 0.34(1) ($x = 0.09$). The reduction of $T_N$ and increase in $\beta$ on addition of induced-moment impurities ($Fe^{2+}$) is less pronounced.

Introduction

Recent theoretical [1-3] and experimental [5-7] work has revealed that antiferromagnetically coupled Heisenberg or XY-type moments on a triangular lattice show a new type of magnetic ordering. In addition to the three-sublattice 120° antiferromagnetic order, a staggered chirality order may be defined. The chirality of an individual triangular plaquette is defined as the sense of rotation of the moments around its edges. This is believed to create new universality classes, in which the critical exponent of the magnetisation, $\beta$, for stacks of such planes is 0.25 and 0.22 for the Heisenberg and XY cases respectively [3, 4]. Recent measurements on CsMnBr₃ and CsVX₃ ($X = Cl, Br$) [5, 7], which are pseudo one-dimensional $XY$ antiferromagnets with interchain exchange pathways forming a triangular array in the $ab$ plane, showed a value of 0.22 ± 0.03 for $\beta$. In addition, being frustrated magnets these materials are susceptible to the formation of canted local states [8] on dilution with diamagnetic impurities. Near such impurities the balance of forces that produces the frustrated ground state may be upset and some of the frustration released by a rotation of neighbouring moments [9].

In this paper we describe the effect on the magnetic ordering processes of CsMnBr₃ of doping with diamagnetic ($Mg^{2+}$) and induced-moment impurities ($Fe^{2+}$) concerning the disruption of the triangular symmetry that leads to staggered chirality ordering, the relative reduction of $T_N$ on doping both types of impurities and the nature of the diffuse magnetic scattering in the region of the magnetic Bragg peaks [10].

Experimental

Single crystals of CsMn$_{1-x}$Mg$_x$Br₃ ($x = 0.05$, 0.09) and CsMn$_{1-x}$Fe$_x$Br₃ ($x = 0.05$, 0.11) were grown by the Bridgman technique using a Crystalox twin-zone furnace. The neutron scattering measurements were conducted at the Institute Laue-Langevin using the triple-axis instrument D10 using the analyser set to zero-energy transfer, wavelength $\lambda = 2.36$ Å. The crystals were aligned with the [002] and [110] directions in the horizontal scattering plane. The scattering intensity was also measured from (001) to (111) above and below $T_N$ to elucidate the nature of the magnetic correlation in the $ab$-plane.

Results and discussion

The scattering intensity centred at (1/3, 1/3, 1) and (2/3, 2/3, 1) was fitted to a sum of a Gaussian function and a linear background term. $\beta$ was derived by fitting the area of this Gaussian, $I(T)$, to the expression

$$I(T) = A e^{2\beta}$$

where $\varepsilon$ is the reduced temperature, $(T_N - T) / T_N$, and takes values from 0.25 to 0.005. The values of $T_N(x) / T_N(0)$ are plotted against $x$ in figure 1, and compared with the predictions of the Hone model [10] for diamagnetic and magnetic impurities in a collection of Heisenberg chains coupled by a molecular field. The exchange constants for Mn-Mn, Mn-Fe and Fe-Fe, were taken from [11], and the size of the moment on $Fe^{2+}$ assumed to be 1.0. As expected and supported by the Hone model, $Fe^{2+}$ impurities, which retain exchange links within the chain are seen to reduced $T_N$ less than $Mg^{2+}$ impurities which sever the chains completely. The dependence of $\beta$ on composition is given in table I. On doping with $Mg^{2+}$ there is a change
At temperatures below $T_N$ we observe strong elastic diffuse magnetic scattering centred at the positions of the magnetic Bragg peaks. This scattering was seen in both the pure and the doped compounds and persisted to temperatures well in excess of $T_N$, being much weaker but still visible at $T = \frac{3}{2}T_N$.

The lineshape of this diffuse component was clearly asymmetric along the (110) reciprocal lattice direction, and will be discussed in more detail elsewhere [13].

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**Table I.** The dependence of the reduced ordering temperature, $T_N(x)/T_N(0)$, and $\beta$ on the type and concentration, $x$, of the impurity.

<table>
<thead>
<tr>
<th>A$^{2+}$</th>
<th>$x$</th>
<th>$T_N(x)/T_N(0)$</th>
<th>$\beta$</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mn</td>
<td>1</td>
<td>0.22</td>
<td></td>
<td>[5]</td>
</tr>
<tr>
<td>Mn</td>
<td>1</td>
<td>0.19</td>
<td></td>
<td>[13]</td>
</tr>
<tr>
<td>Mg</td>
<td>0.018</td>
<td>0.745</td>
<td></td>
<td>[12]</td>
</tr>
<tr>
<td>Mg</td>
<td>0.048</td>
<td>0.531</td>
<td></td>
<td>[12]</td>
</tr>
<tr>
<td>Mg</td>
<td>0.050</td>
<td>0.525 (5)</td>
<td>0.31 (1)</td>
<td>[12]</td>
</tr>
<tr>
<td>Mg</td>
<td>0.090</td>
<td>0.262 (2)</td>
<td>0.34 (1)</td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>0.050</td>
<td>0.740 (5)</td>
<td>0.22 (1)</td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>0.110</td>
<td>0.600 (5)</td>
<td>0.24 (1)</td>
<td></td>
</tr>
</tbody>
</table>

in $\beta$ from a value close to that predicted for the new universality classes towards that of the 3-D Heisenberg (0.35) or $XY$ (0.33) ones, as might be expected on disruption of the frustrated triangular ground state and the 3-D arrangement of the combined triangular $ab$-planes. The values of $\beta$ for Fe$^{2+}$-doped CsMnBr$_3$ showed that this type of doping does not disrupt the chirality ordering or its 3-D character.

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