On a new type of sublattice tipping in noncollinear antiferromagnets

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An explanation is offered for the 30° tipping of sublattices which has been observed in the noncollinear antiferromagnet $CsMnI_3$. In contrast to the ordinary 90° spin flop, this phase transition is due to nonlinear and relativistic corrections to the susceptibility. [S0021-3640(98)01223-7]

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Tipping of sublattices (see, for example, Ref. 1) in collinear antiferromagnets is associated with the anisotropy of the magnetic susceptibility. In easy-axis antiferromagnets in a critical magnetic field directed along a symmetry axis, the antiferromagnetic vector rotates by 90°, and the maximum of the susceptibility tensor is oriented in the direction of the field. A transition of this type can also be observed in noncollinear magnets with sufficiently low exchange symmetry, such that anisotropy of the magnetic susceptibility is present.

In Ref. 2 it was found that in the noncollinear antiferromagnet $CsMnI_3$, when the magnetic field reaches a certain value less than the ordinary spin flop field, the orientation of the sublattices changes abruptly by 30° (see Fig. 1). The axial magnetic susceptibility tensor in $CsMnI_3$ does not change with this spin rotation. Thus an unexpected reorientation phenomenon was observed in Ref. 2. To describe this new type of sublattice tipping the nonlinear and relativistic corrections to the magnetic susceptibility must be taken into account.

The structure of antiferromagnetic $CsMnI_3$, according to the theory of exchange symmetry,[3] is determined by two unit orthogonal spin vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ (cf. with Ref. 4):

$$\mathbf{S}\sim\mathbf{A}\exp(i\mathbf{Q}\cdot\mathbf{r})+\mathbf{A}^*\exp(-i\mathbf{Q}\cdot\mathbf{r}),\quad \mathbf{A}=\mathbf{l}_1+i\mathbf{l}_2,\quad \mathbf{Q}=\left(\frac{4\pi}{3a},0,\frac{\pi}{c}\right). \tag{1}$$

The anisotropy energy of first order in $(v/c)^2$ reduces to the single invariant $(\beta/2)n_z^2$, $\mathbf{n}=\mathbf{l}_1\times\mathbf{l}_2$. For $\beta>0$ the vector $\mathbf{n}$ is perpendicular to the C_6 symmetry axis (z axis) of the crystal. In the presence of a magnetic field the orientation of the vector $\mathbf{n}$ is determined by minimizing the energy

$$-\frac{\chi_\parallel-\chi_\perp}{2}(\mathbf{n}\cdot\mathbf{H})^2+\frac{\beta n_z^2}{2}, \tag{2}$$

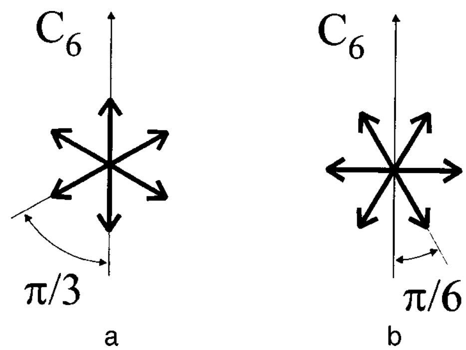

FIG. 1.

where $\chi_\parallel$ ($\parallel\mathbf{n}$) and $\chi_\perp$ ($\perp\mathbf{n}$) are the magnetic susceptibilities, and in $CsMnI_3$ $\chi_\parallel > \chi_\perp$. If the field $\mathbf{H}$ is directed along a hexagonal axis, then for

$$H < H_c = \sqrt{\frac{\beta}{\chi_\parallel - \chi_\perp}} \tag{3}$$

the vector $\mathbf{n}$ lies in the basal plane, and for $H > H_c$ (H_c is the ordinary spin flop field) the vector $\mathbf{n}$ is parallel to the $\mathbf{z}$ axis.

The orientation of $\mathbf{n}$ in the basal plane and the orientation of the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ in the spin plane are determined by invariants of sixth order in the components of the vector $\mathbf{A}$ (see Ref. 5):

$$I_1 = A_z^6 + A_z^{*6}, \quad I_2 = (A_x + iA_y)^6 + (A_x - iA_y)^6 + (A_x^* + iA_y^*)^6 + (A_x^* - iA_y^*)^6. \tag{4}$$

AFMR and NMR investigations[6,7] show that the anisotropy I_2 is small. Therefore we neglect its contribution to the energy.

In a magnetic field, three additional invariants must be taken into account:[5]

$$I_3 = A_z^4(\mathbf{A}\cdot\mathbf{H})^2 + A_z^{*4}(\mathbf{A}^*\cdot\mathbf{H})^2, \quad I_4 = A_z^2(\mathbf{A}\cdot\mathbf{H})^4 + A_z^{*2}(\mathbf{A}^*\cdot\mathbf{H})^4,$$

$$I_5 = (\mathbf{A}\cdot\mathbf{H})^6 + (\mathbf{A}^*\cdot\mathbf{H})^6. \tag{5}$$

It is evident from the structure of these invariants that I_3 and I_4 are exchange-relativistic and I_5 is purely exchange.

In a field $\mathbf{H}\parallel\mathbf{z}$ less than the ordinary spin flop field (i.e., reorientation of the vector $\mathbf{n}$), the energy has the form

$$f(H)\cos 6\phi, \quad f(H) = b_1 + b_3H^2 + b_4H^4 + b_5H^6, \tag{6}$$

where ϕ is the angle between the vector $\mathbf{l}_1$ and the $\mathbf{z}$ axis, and b_k are the coefficients of the invariants I_k. Here $b_1 \sim (v/c)^6$, $b_3 \sim (v/c)^4$, and $b_4 \sim (v/c)^2$ are relativistic constants, and b_5 is an exchange constant. The expression (6) has two groups each with six equivalent extrema:

$$1)\ \ \phi = \frac{\pi}{3}i, \quad i = 0,..,5; \qquad 2)\ \ \phi = \frac{\pi}{6} + \frac{\pi}{3}i, \quad i = 0,..,5. \tag{7}$$

For $f(H) < 0$ the first group of solutions corresponds to minimum energy (in $CsMnI_3$ $f(0) = b_1 < 0$). When the sign of $f(H)$ changes, the tipping under discussion will occur in a certain critical field H_{c1}, and the second group of solutions will materialize. All terms in the expression for $f(H)$ become of the same order of magnitude in fields $H \sim v/c$ (i.e., in fields of the order of the ordinary spin flop field H_c), and, depending on the values of the constants b_k, the function $f(H)$ can change sign once, twice, or three times with increasing field. In $CsMnI_3$, in fields below H_c a single transition was observed from states of the type 1 (Fig. 1a) to states of the type 2 (Fig. 1b), i.e., the function $f(H)$ changes sign once at a field $H_{c1} \approx 0.7 H_c$.

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