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OBSERVATION OF ADDITIONAL FMR RESONANCES IN A TWO-SUBLATTICE FERROMAGNET

S. L. Hutton and J. E. Drumheller

Department of Physics, Montana State University Bozeman MT 59715 U.S.A.

Abstract. - Theoretical calculations predict additional resonance peaks in two-sublattice ferromagnets of single magnetic entities. Layered magnetic compounds provide possible systems to observe such effects because of weak inter-planar exchange. Theory of two sublattice uniaxial ferromagnets and preliminary inter-planar exchange measurements in \[\text{[NH}_3\text{(CH}_2\text{)}_7\text{NH}_3\] CuBr\(_4\) are presented.

Ferromagnetic resonance [1] (FMR) is well established as a method for obtaining anisotropy fields. Because of the 3-dimensional isotropy of exchange fields in typical ferromagnetic systems, they are normally treated as one-sublattice systems and reference to exchange fields has not been an explicit part of the free energy expansion. There are classes of the layered magnetic compounds [2], however, which offer the possibility of being described as two sublattice ferromagnets. These systems are characterized by a strong intraplanar ferromagnetic exchange field (which determines \(T_c\)) and a weaker inter-planar exchange which may be either ferromagnetic or antiferromagnetic (AF) [3]. Until now, resonance in the collapsed state corresponding to two-sublattice systems have been observed only in the AF compound \((\text{C}_2\text{H}_5\text{NH}_3)_2\text{CuCl}_4\) [4]. In this paper we present a development of FMR in a uniaxial two-sublattice ferromagnet and give preliminary data which support the two-sublattice hypothesis in the layered ferromagnet \([\text{NH}_3\text{(CH}_2\text{)}_7\text{NH}_3\] CuBr\(_4\).

The model is a uniaxial ferromagnet with the external magnetic field applied perpendicular to the easy axis. While this model is overly simple, it will point out the qualitative features to be expected from more complicated systems. In order to develop the theory of two-sublattice FMR, we choose to generalize the theory developed by Baselgia et al. [5] which has the benefit that high-field limiting cases are obtainable by direct substitution of field angles for magnetization angles. If we consider a system with the external field \((H)\) applied along the \(z\)-axis with an uniaxial anisotropy field along the \(x\)-axis, an appropriate free energy model is

\[
F = (1/2) K \left[ (M_x^1)^2 + (M_x^2)^2 \right] - H \left[ (M_x^1 + M_x^2) + \varepsilon [M^1 \cdot M^2] \right] \tag{1}
\]

where \(M^1\) and \(M^2\) are the sublattice magnetizations. Since we wish the easy axis to lie along the \(z\)-axis, \(K\) is negative and since the inter-planar exchange is ferromagnetic, \(\varepsilon\) is negative.

In the theory of Baselgia et al. [5], the static equilibrium orientations of \(M^1\) and \(M^2\) are obtained by minimizing the free energy with respect to the two body Cartesian components of magnetization, namely, \(M_1\) and \(M_2\) with \(M_3\) parallel to \(M\). For the model (Eq. (1)), the two equations which yield the equilibrium angles are

\[
\begin{align*}
\frac{\partial F}{\partial M_x^1}|_{M_x^1=0, M_x^2=0, M_z^1=M_z^2} &= 0 \\
\frac{\partial F}{\partial M_x^2}|_{M_x^1=0, M_x^2=0, M_z^1=M_z^2} &= 0
\end{align*} \tag{2}
\]

and

\[
\frac{\partial F}{\partial M_z^1}|_{M_x^1=0, M_x^2=0, M_z^1=M_z^2} = 0 \tag{3}
\]

where the sublattice index \(\sigma\) refers to sublattice 1 or sublattice 2. For this model, equation (3) gives the solution for \(\phi\):

\[
-KM_s \sin \theta \sin \phi \cos \phi = 0 \tag{4}
\]

where \(M_s\) is the sublattice saturation magnetization. Since we seek the minimum in the free energy, the correct choice for \(\phi\) will be \(\phi = 0\). The solution for \(\theta\) is \(\theta = 0\) if \(|H/KM_s| > 1\) and \(\cos \theta = -H/KM_s\) otherwise.

The resonance condition for a two-sublattice ordered state system is obtained by generalizing the torque equations are for each sublattice:

\[
\frac{dM_{\text{body}}^\sigma}{dt} = \gamma M_{\text{body}}^\sigma \times H^\sigma \tag{5}
\]

where \(H^\sigma\) is the effective field acting on sublattice \(\sigma\) and is given by

\[
H^\sigma = -\sum_{\sigma=1}^{2} \frac{\partial F}{\partial M_{\text{body}}^\sigma}. \tag{6}
\]

By expanding the right hand side of equation (5) in a Taylor series for \(M_{\text{body}}^\sigma\) small quantities and assuming harmonic time dependence for \(M_{\text{body}}^\sigma\) and \(M_{\text{body}}^\sigma\), we obtain the resonance equation (6) for two-sublattice FMR:

\[
(\omega / \gamma)^2 = \left[ M_s F M_{\text{body}}^1 M_{\text{body}}^1 - F M_{\text{body}}^2 \right] \pm M_s F M_{\text{body}}^1 M_{\text{body}}^2 \tag{7}
\]
where $F_{M_1 M_2} = \frac{\partial^2 F}{\partial M_1^2 \partial M_2^2}$ for example. With our model, we have (using the solution $\phi = 0$):

$$F_{M_1 M_2} = \frac{K \cos^2 \theta, F_{M_1} = K M_s \sin^2 \theta - H \cos \theta - \epsilon M_s, \quad F_{M_1 M_2} = F_{M_1} - F_{M_2} = \epsilon.$$ (8)

In the low field region, where $|H / K M_s| < 1$, we then have the two low field solutions:

$$(\omega / \gamma)^2 = (K M_s + \epsilon M_s (1 \pm 1))^2 - [1 + (\epsilon M_s / K M_s) (1 \pm 1)] H^2$$ (9)

and in the high field region where $|H / K M_s| > 1$ the two solutions are given by:

$$(\omega / \gamma)^2 = [H - \epsilon M_s (1 \pm 1)][H - \epsilon M_s (1 \pm 1) + K M_s]$$ (10)

where the signs in equations (9, 10) are correlated. A plot of the these frequencies for two values of $\epsilon$ is shown in figure 1. From the intersections of the line $\omega = \text{const.}$ with the resonance curves it is obvious that one can observe up to four resonances. From equations (5, 10) it is straightforward to show that the modes correspond to in-phase and 180° out-of-phase precession of the two sublattices about their static equilibrium orientations (see Fig. 1).

Fig. 1. — Theoretical dependence of the two resonance modes on magnetic field for two values of $\epsilon$. The usual FMR curve corresponding to mode a (solid line) remains unchanged in both cases while the other curve corresponding to mode b (broken line) moves to higher frequencies. Resonance fields observed from ESR apparatus are determined by the intersection of the curve $\omega = \text{const.}$ with the resonance curve.

Preliminary evaluation of FMR data indicates that a more complicated model is necessary for the compound $[\text{NH}_3(\text{CH}_2)_7\text{NH}_3]\text{CuBr}_4$. This would likely include uniaxial anisotropy fields along two axes and possibly a small second order anisotropy field. In addition, it may be necessary to introduce anisotropy into the inter-planar exchange since the resonance peaks have been seen to vary in their separation as a function of angle. Initial data fits indicate that at 4 K, the average intersublattice exchange field is 0.05 K. Susceptibility measurements [3] have shown that the inter-planar exchange field is < 1 K and powder susceptibility measurements of neighboring AF compounds have given an estimate for $J$ between 0.1 K and 0.05 K.

In addition the relative intensity variation of our resonances is roughly $\cos^2(2\theta_H)$ where $\theta_H$ is the field polar angle in the plane perpendicular to the easy axis (see Fig. 2). This is analogous to the intensity variation observed by Reimann et al. [4] in the collapsed state of the AF system $(\text{C}_2\text{H}_2\text{NH}_3)_2\text{CuCl}_4$. The system for this work shows this intensity variation associated with the resonance which is lower in field owing to the opposite sign of the exchange.

Fig. 2. — Relative intensity of the two high-field resonances. For comparison, the function $3 + 14 \cos^2(2\theta_H)$ is shown. The magnetization vector is not necessarily parallel to the field angle, the intensity data are not at minimum at $\theta_H = 45°$.

Determination of inter-planar exchange in the layered magnetic compounds is possible with standard ESR apparatus for layered ferromagnetic compounds. In the compound $[\text{NH}_3(\text{CH}_2)_7\text{NH}_3]\text{CuBr}_4$, measurement of inter-planar exchange field is in good agreement with estimates obtained from susceptibility measurements.

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[6] Equation (7) is not valid for systems which have a Dzyaloshinsky-Moriya interaction as part of their free energy expansion.