MÖSSBAUER STUDIES OF SPIN REORIENTATIONS IN OXIDES

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Mössbauer spectroscopy has been remarkably useful in understanding several properties of the mixed oxides. The investigations relating to spin arrangements form an important part of the successful applications of this method. In mixed spinel ferrites and garnets, Mössbauer spectroscopy unambiguously showed the non-collinearity of spins in the same sublattice in several cases. The detailed investigations showed features predicted by the localized canting model (LCM), viz., the simultaneous presence of non-collinearity at the two non-equivalent sites in certain cases, 'spin reversal' of ions without magnetic ions on the nearest non-equivalent sites. However, it has not been possible to observe other details of the model. Another striking result obtained is that non-collinearity decreases rapidly as the temperature increases, disappears at temperatures below 80 K. These studies have not, however, succeeded in determining the exchange constants unambiguously. In orthoferrites, the complications due to the variations in the environments of ions in the same sublattice are not present. Consequently, studies of spin reorientations (SR) relative to the crystal axes and other magnetic properties have provided accurate results, ideal for theoretical analyses. SR is due to the anisotropic-symmetric and antisymmetric exchange interactions between the rare earths and iron group ions, which are much smaller than the isotropic part of the exchange interaction. Thus, a detailed theoretical analysis expresses SR parameters in terms of these smaller interactions and the external magnetic field, if present. The number of parameters involved is, however, large. Analyses of the experimental data are made using a simpler two-sublattice model involving a smaller number of parameters. This, nevertheless, makes comparison between similar orthoferrites possible. There are other oxides as well which show SRs due to changes in the signs and magnitudes of the crystal field anisotropic constants. Such studies are useful for investigating crystal field interactions.

1. Introduction

Exchange interactions among magnetic ions in solids, though the basic cause of magnetic ordering, have not been predicted accurately from theory due to the well-known complexities. They have been obtained mainly from experimental studies. Mössbauer spectroscopy can provide a convenient method for obtaining information about these constants through spin orientation studies, precisely in certain circumstances.

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Spin orientation studies can be classified into three groups. In the first class, mutual spin orientations of ions in the same sublattice are investigated. These studies have been mainly concerned with mixed spinel ferrites and garnets. Early developments in theory [1—7] were mainly directed to the interpretation of the magnetization of these diamagnetically substituted oxides. Early experimental studies using neutron diffraction [8,9] and Mössbauer spectroscopy [10] substantiated the presence of non-collinearity. The realistic theoretical treatment of the problem, taking into consideration the statistical fluctuations in the environments of the magnetic ions in a sublattice, was given by Rosencwaig [11]. Subsequent Mössbauer studies provided detailed information about the non-collinearity and are described in sect. 2.

In the other classes of investigations, the dependences of the spin orientations relative to crystalline axes on temperature and external magnetic field are determined. Extensive studies have been made on SRs in orthoferrites, RFeO₃. Unlike the first category, in which the interplay of the isotropic parts of exchange interactions is responsible for the non-collinearity, SR relative to the crystalline axes in orthoferrites is mainly due to the weak anisotropic-symmetric and antisymmetric exchange forces. The environments of all the magnetic ions in any sublattice are identical. The theory given by Yamaguchi [12], however, highlights the complexities involved in obtaining information from the unambiguous experimental results. Section 3 discusses these experimental results.

Lastly, SRs relative to the crystalline axes mainly due to the changes in signs and magnitudes of the crystal field anisotropic constants, found in other oxides such as SrTb₂Fe₂O₇, α-Fe₂O₃, Y₃Fe₅₋ₓSiₓO₁₂, SrGdFeO₄, etc., are summarized in sect. 4. There are a number of interesting spin orientation studies in metallic substances as well, but they have not been included in this article.

2. **Non-collinear spin structure in spinel ferrites and garnets**

When diamagnetic cations are substituted for magnetic ions in a sublattice of a spinel ferrite or garnet, the magnetic ions in the neighbouring sublattice split into two mutually oriented magnetic sublattices. The study of such non-collinearity can provide information about the intersublattice and intrasublattice exchange constants, and has provided interpretation [13] of the anomalous dependence of the magnetization of these diamagnetically substituted oxides on the concentrations of the cations.

2.1. **YAFET—KITTEL THEORY**

We shall consider spinel ferrite in the discussion of the theoretical aspects of non-collinear spin configurations in oxides. The non-collinearity of spins found in garnets can be understood using similar considerations. The unit cell of the spinel structure containing eight molecules can be divided into octants, fig. 1. Oxygen