MAGNETIC STRUCTURE OF DISORDERED Au-Mn ALLOYS

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Résumé. — Nous discutons ici la structure hyperfine Mössbauer de Au¹⁹⁷ dans les alliages Au-Mn comprenant moins de 20 % Mn. Nous avons développé un modèle, conceptuellement simple, qui réussit à expliquer la structure hyperfine compliquée obtenue pour l'alliage à 20 % Mn. Avec des paramètres réduits par une échelle appropriée, la structure hyperfine obtenue est bien prédite pour les alliages comprenant moins de Mn. Des calculs fondés sur d'autres modèles plausibles ne reproduisent pas les spectres observés.

Abstract. — This article discusses the Mössbauer hfs of Au¹⁹⁷ in magnetically ordered crystallographically disordered Au-Mn alloys containing up to 20 % Mn. A conceptually simple model is developed which is successful in explaining the complex hfs observed in the 20 % alloy. With appropriately scaled parameters, the hfs observed in alloys with smaller Mn concentrations is also predicted. Calculations based on a number of other plausible models fail to reproduce the observed hfs.

Previous studies by Meyer [1] of the susceptibility and magnetization of the crystallographically ordered (bcc) and disordered (random substitutional fcc) phases of Au₄Mn have shown that both have a susceptibility of about 4.8 μB per Mn ion above their magnetic ordering points. The ordered phase of Au₄Mn has a low temperature magnetization of ~ 4.2 μB per Mn, corresponding to the ferromagnetic alignment of the Mn ions, but the disordered phase shows a much smaller magnetization [1]. This suggests that the magnetic properties of the Au-Mn systems arise from local moments on the Mn ions, but that antiferromagnetic coupling among the Mn ions can reduce the net moment of the disordered system. This is not surprising, since the Ruderman-Kittel oscillating spin density mechanism provides an exchange interaction which changes sign with distance. A wide range of magnetic structures appears in Au-Mn intermetallic compounds [2, 3, 4, 5, 6, 7] showing a strong correlation with the Mn-Mn nearest neighbor distance (see Table I).

<table>
<thead>
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<th>Table I</th>
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<td>Mn distances and couplings in ordered Au-Mn intermetallics</td>
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<td><strong>Formal</strong></td>
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The Mössbauer spectra of Au¹⁹⁷ in the ordered Au₄Mn shows a well resolved hyperfine spectrum [8, 9], arising from an effective field, H₄Mn, at the gold nucleus, coming from the contact interaction with spin-polarized conduction electrons. The hfs of the disordered phase material, though, shows a broad spectrum with the lines only poorly resolved. This presumably results from the range of environments for the gold atoms, caused by the random alloy statistics. It would at first appear to be almost impossible to construct a microscopic model describing the origins of the hfs in this random alloy. However, with the aid of a number of assumptions described below, it is possible to develop a model which reproduces the observed hfs very closely. The successful duplication of the experimental hfs is not a guarantee that the model is correct. However, calculations based on a number of other plausible models yield quite different results from those obtained experimentally. The success of the model in predicting the observed hfs in other (random fcc) alloys with less Mn also suggests that the calculation described below does describe the microscopic magnetic structure of these materials.

We make the following assumptions:

1. We assume that the alloys are random, with no short range order.
2. The isomer shift (IS) is assumed to be increased by 0.63 mm/s by each Mn NN to a gold (this value is determined from the well resolved spectrum of the ordered phase Au₄Mn).
3. The hf field, H₄Mn, at a gold atom is assumed to come from the linear superposition of H₄Mn, from the Mn NN's, and H₄Mn, a «long-range» term which lump together the effects of all the magnetic ions outside the NN shell.
4. Quadrupole hfs is neglected. (This is suggested by the negligible quadrupole coupling [8] in ordered Au₄Mn and in ordered and disordered [8] Au₄V.)
5. Each Mn ion contributes a fixed amount, H₄Mn, to H₄Mn, but it can either add to or subtract from H₄Mn depending on its spin direction.

The data in Table I suggest that the coupling between Mn ions is antiferromagnetic if they are less than ~ 3.2 Å apart and ferromagnetic if they are further apart [4]. Applying this picture to a sample cell of the fcc structure (Fig. 1) shows that if two Mn ions are NN's, they should be antiferromagnetically coupled, and if they are further apart, but still within the same cell, they should be coupled with spins parallel. The application of this algorithm leads to a self-consistent spin configuration for each possible local arrangement of Mn ions.

The calculation of the hfs on this model proceeds by the following stages: First, for each possible number of Mn ions in the cell shown, the spin configuration is
worked out for various possible positions of the Mn ions. This calculation is facilitated by the use of equivalence classes and a simplified Monte-Carlo approach. For 4 Mn nearest neighbors to the gold, for example, $H_{\text{iso}}$ can be 0, 2, or $4 \times H_{\text{NN}}$, with a probability depending on the (random) distribution of Mn ions at the 12 possible sites. The hfs for the gold « test atom » shown at the center of the cell in figure 1 is then calculated by using an IS determined from the total number of Mn nearest neighbors, and $H_{\text{int}} = H_{\text{dist}} + H_{\text{iso}}$. Because the number of Mn NN’s is determined by random alloy statistics, and the $H_{\text{iso}}$ for a particular number of Mn ions can also take on a number of values, the calculation is complex, but straightforward. Because the IS per Mn NN can be estimated from the ordered phase Au$_4$Mn, the only free parameters in the model are $H_{\text{iso}}$ and $H_{\text{dist}}$; the remainder of the calculation is determined by the probability distributions.

The calculated hfs, shown by the solid line in figure 2, is in good agreement with the observed spectrum for the 20 % alloy, with the parameter values $H_{\text{iso}} = 105$ kOe and $H_{\text{dist}} = 425$ kOe. To calculate the spectra for the other alloys, the model requires that the IS and $H_{\text{iso}}$ values should stay the same, and that only the $H_{\text{dist}}$ values should be changes. A severe test of the model’s validity is to determine $H_{\text{dist}}$ for alloys with < 20 % Mn from the value providing the best fit to the data for the 20 % alloys, taking account of the reduced concentration of Mn ions, and the relatively greater proportion of ferromagnetically coupled Mn ions. This leads to the result for 15, 10, and 3.5 % Mn $H_{\text{dist}} = 350, 260, 110$ kOe, and those values have been used to calculate the solid lines in figure 2 (Somewhat better agreement with the experimental hfs can be obtained with slightly smaller values of $H_{\text{dist}}$.)

The success of the theory, with no arbitrary adjustable parameters, in fitting the observed spectra for the more dilute alloys suggests that the model on which the calculations is based is a meaningful description of the sources of the hfs in the Au-Mn disordered alloys.

References

[6] Ferromagnetic coupling between Mn ions lying 3.31 Å apart on square planar lattice. Overall magnetic structure is flat spiral. Herpin (A.), Meriel (P.) and Villain (J.), Compt. Rend., 1959, 249, 1334.