Evidence against Metastable Phase Separation in *Au*Fe Alloys

In a recent Letter, Violet and Borg[1] (VB) have presented an analysis of Mössbauer (ME) spectra of the metastable quenched alloy $Au_{1-x}Fe_x$ ($0.105 < x < 0.33$). In this concentration range a double magnetic transition has previously been observed[2]: a paramagnetic-ferromagnetic transition at T_c, followed at a lower temperature T_f to a spin-glass[2] or spin-glass-like[3] state. In contrast to this picture VB explain their (4.2 K) results on the basis of the known short-range order in $Au_{1-x}Fe_x$, postulating a two-*phase* model: a low-hyperfine-field (H_{HF}) component from the Au-rich solid-solution matrix (low-field phase), and a high-H_{HF} component (high-field phase) from the Fe-rich platelets (discovered by Dartyge, Bouchiat, and Monod[4] in x-ray measurements). They claim that this model may explain the observed double transition.

Our first comment is that within this model a completely different temperature dependence of ME spectra is expected as compared to that actually observed: For $T \gtrsim T_f$ the solid-solution matrix must become paramagentic, and it would be expected that the remaining Fe platelets would become superparamagnetic. Figure 1 shows spectra for $Au_{0.832}Fe_{0.168}$ below and above $T_f \sim 45$ K: The analysis of the hyperfine field distribution $P(H_{HF})$ shows no zero-H_{HF} (paramagnetic) component appearing at these temperatures. However, since in the VB model the fraction of Fe atoms in the low-field phase is 65% for $Au_{0.832}Fe_{0.168}$ a strong (central) paramagnetic (quadrupole-split) spectral component should appear for $T \gtrsim T_f$ contributing $\sim 65\%$ to the total spectral area. Figure 1 shows unambiguously that this is not the case. Our observation rules out the model of VB. Also our $P(H_{HF})$ analysis for $Au_{0.832}Fe_{0.168}$ at 4.2 K in external fields has shown[5] that the local response to applied fields is *homogeneous* for all Fe atoms, irrespective of the local Fe concentration, in contradiction to a two-phase interpretation.

The model of VB also does not explain the two most important and general properties below T_f: a spontaneous canting of the moment directions, and an associated anomalous increase in the average H_{HF} and local moment ($S \propto \bar{H}_{HF}$) as T is lowered. It was shown[3,5,6] that these properties are observed not only in *Au*Fe but in many different double-transition systems which are known to be random (e.g., amorphous Fe-Ni and Fe-Mn alloys, and $Mg_{1+t}Fe_{2-2t}Ti_tO_4$). The results show[3,6] that the appropriate order parameter in the low-temperature state is the transverse spin component S_t.

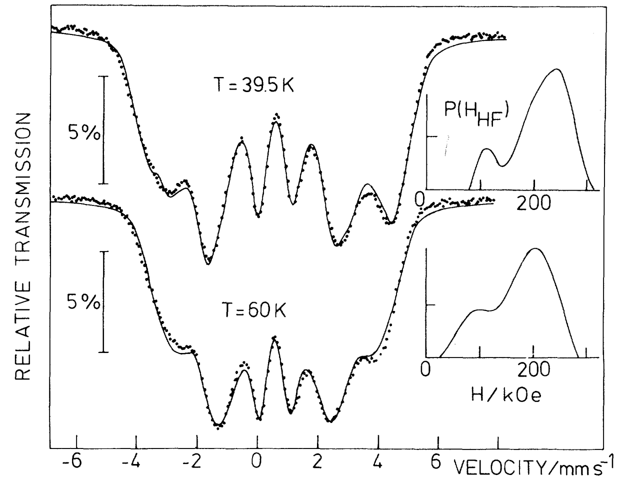

FIG. 1. ^{57}Fe Mössbauer spectra of 16.8-at.% Fe-Au (quenched) at $T = 39.5$ and 60 K measured in zero external field. Insets: Hyperfine field distribution $P(H_{HF})$ for each spectrum as obtained by least-squares fitting.

We would like to thank Dr. J. Lauer for the use of unpublished data.

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Received 4 November 1983
PACS numbers: 64.75.+g, 61.55.Hg, 76.80.+y, 81.30.Mh

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