

**Violet and Borg Respond:** Because of space limitations we can respond to only three criticisms of Monod and Campbell<sup>1</sup> (MC) and Brand and Keune<sup>2</sup> (BK).

MC (1): The Au-Fe system cannot be represented "as a strict two-phase system." Reply: We used the terms "phase" and "environment" interchangeably, implying that we do not consider the two iron "environments" to be strict phases in the classical meaning.<sup>3</sup> We reiterate that our Mössbauer spectra, which agree with those of other investigators,<sup>4-6</sup> are well described by two superposed six-line magnetic hyperfine spectra (MHS). Curiously, MC and BK object to this analysis but make no mention of or objection to our supporting evidence, viz. the back calculation of the bulk composition,  $X_c$ , from the Mössbauer data. Our spectra could also be described by the methods of Ridout<sup>5</sup> and Window.<sup>6</sup> By including enough parameters (in this case, near-neighbor shells) any spectrum, of course, can be described. However, we maintain our hypothesis that the Fe in these Au-Fe alloys resides predominantly in two distinct "chemical environments." We further hypothesize that within the broad family of short-range (SRO) configurations associated with  $(1, \frac{1}{2}, 0)$  diffuse scattering there are predominantly two such configurations in these Au-Fe alloys corresponding to the two environments which give rise to the two superposed MHS. Except for the 33-at.% alloy, the two superposed MHS for each of the other alloys collapse with increasing temperature, merge, and become an asymmetric doublet at  $T_0$ , the upper ordering temperature.<sup>7</sup> Also, the upper ordering temperatures, determined from the temperature variation of either of the two superposed MHS individually, agree within a few degrees. Conversely, the 33 at.% alloy consists of  $\text{Au}_{0.73}\text{Fe}_{0.27}$  (approximate composition) and  $\alpha$ -Fe, each with widely differing ordering temperatures, 370 and 1043 K, respectively.<sup>7</sup> After completing the initial results contained in our Letter, we initiated a transmission electron-microscopy study of a 17- and a 25-at.% alloy at the National Center for Electron Microscopy, Lawrence Berkeley Laboratory.<sup>8</sup> This study<sup>9</sup> failed to reveal even platelets, much less precipitates, in excellent agreement with our previous transmission electron-microscopy investigation<sup>10</sup> of a 17-at.% alloy, although diffuse scattering was observed.<sup>9</sup> These results imply that the platelet model of Dartyge, Bouchiat, and Monod<sup>11</sup> is wrong. It also means that our identification of the high-field and low-field "phases" with the platelet and matrix structures,<sup>11</sup> respectively, is perforce wrong.

MC (2): The Au-Fe spectra cannot be fitted with two superposed MHS because Window could not do so. Reply: We object to the supposition that, since Window "could not fit his spectra with two six-line patterns only," it follows that we cannot do so. We have reexamined Window's paper and find no mention of the possibility of fitting his spectra with two MHS nor any attempt to do so. While Window's work, as well as Ridout's, show that these spectra can be fitted by including enough near-neighbor shells (Window took three and Ridout took five), Window's and Ridout's work suffers from at least two problems: (a) They assumed a random solid solution, whereas short-range order is now known to exist,<sup>12</sup> and (b) they used the 3:2:1:1:2:3 constraint in their least-squares fitting. This constraint, when applied to these Au-Fe alloys, leads in general to poor fits. Much better agreement is obtained by use of the less stringent symmetry constraint or the 3:-:1:1:-:3 constraint. Lauer and Keune<sup>13</sup> extracted a distribution of hyperfine fields from their analysis of the MHS of a 16.8-at.% alloy at 4.2 K. Their distribution consists of *two peaks* centered at  $H$  values that are in excellent agreement with the values we quote for our 16.8 at.% alloy. They "attribute the high-field part (of their distribution)... to Fe-rich clusters, while the low-field part... is associated with Au-rich clusters." If we ignore for the moment the detailed meanings of their "clusters" and our "chemical environments," both interpretations in effect postulate two distinct chemical environments. However, except for our 25- and 33-at.% alloys, the high-field environments in our alloys turn out to be *Fe-enriched* rather than Fe-rich, while the low-field environments are always Au-enriched.

MC (3): "Short-range order in CuMn is toward *anticlustering* and so is just the opposite to that of AuFe." Reply: Cu-Mn and Au-Fe exhibit the same  $(1, \frac{1}{2}, 0)$  diffuse scattering. On the basis of the work of Cable *et al.*,<sup>14</sup> it is not unreasonable to suppose that, of the many configurations associated with this SRO, the SRO in Cu-Mn might also involve predominantly two configurations. The word "anticlustering" aggravates an already confused situation since it implies random aggregations of similar or dissimilar nearest neighbors.

Finally, the idea of spontaneous "canting" in our opinion is not only confusing but fundamentally incorrect. In its original meaning, this term referred to "canted antiferromagnetism, in which a single-axis antiferromagnetic structure is modified by having the moments *deviate slightly* from the axis"<sup>15</sup> (our italics). We know of no evidence for spin canting *per se* in Au-Fe in the customary sense of

this term. In the so-called "canting transition" the vast majority of moments may very well continue to lie near the axis (field direction) rather than along a so-called "canting angle." As  $T$  decreases below the lower ordering temperature,  $T_f$ , the moment most likely changes from a fairly unique to an increasingly distributed quantity making large deviations from the axis increasingly probable. The extent of this distribution is a function of *composition*, *temperature*, and *field*. The general trend of the magnetic behavior, regardless of composition, shows that the sample is easily polarized in the temperature range just below  $T_0$  as would be expected from general considerations of cooperative phenomena. Below  $T_f$  there is a conversion from correlated spin alignment toward an overall random (spin-glass) spin orientation at lower temperatures. The average spin orientation at each point is adequately described by the usual spin correlation functions,<sup>16</sup> but the overall macroscopic moment so calculated from Mössbauer data is less than that measured by a conventional magnetometer. Moreover, the rate at which a more or less polarized spin array relaxes into a spin-glass is temperature dependent. A thorough discussion of these phenomena for several compositions, including Mössbauer polarization and magnetization measurements, has been given by Borg and Kitchens.<sup>16</sup> The presence of SRO apparently has little if any influence on the appearance of these phenomena.

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