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Effects of Fe on the ferromagnetic properties of Al-Mn-based quasicrystals

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Magnetization and Mössbauer-effect studies of ferromagnetic Al-Mn-Si and Al-Mn-Cu-Ge quasicrystals are presented. All alloys show paramagnetic or spin-glass behavior in addition to ferromagnetism at low temperatures. The effects of Fe substitution on the magnetic properties were investigated. Possible models for ferromagnetic order in quasicrystals are discussed.

I. INTRODUCTION

Recent interest in the magnetic properties of quasicrystalline materials has been generated by reports of ferromagnetic order in icosahedral Al-Ce-Fe,¹ Al-Mn-Ge,² and Al-Mn-Si (Ref. 3) alloys. These alloys show Curie temperatures which range from ~ 100 to ~ 500 K and relatively low saturation magnetizations. It has been suggested that a model involving weak itinerant ferromagnetism or a noncollinear spin arrangement may be applicable to these materials⁴ although, as yet, concrete evidence for either has not been presented. Perhaps one of the most interesting features of these ferromagnets is the apparent superposition of ferromagnetism and spin-glass behavior. Recent ferromagnetic resonance studies of ferromagnetic Al-Mn-Si have provided compelling evidence that both behaviors are characteristic of the same Mn moments.⁵ Certainly there is still much to be learned in order to fully understand the magnetic behavior of these interesting materials. In the present work we report on Mössbauer-effect and magnetic-susceptibility studies of ferromagnetic Al-Mn-Si and Al-Mn-Fe-Cu-Ge quasicrystals.

II. EXPERIMENTAL METHODS

Alloys of Al-Mn-Si and Al-Mn-Fe-Cu-Ge were prepared by arc melting high-purity components and melt spinning onto a single Cu roller with a surface velocity of 60 m/s. X-ray diffraction measurements indicated that all alloys reported in this work were essentially single-phase icosahedral quasicrystals.

Mössbauer-effect measurements were made using a Pd ⁵⁷Co source and a Wissel System II spectrometer. Magnetic-susceptibility measurements were made with a SHE superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS

A. Al-Mn-Si alloys

Dunlap *et al.*³ have shown that high Si content quasicrystals of the composition $\text{Al}_{80-x}\text{Mn}_{20}\text{Si}_x$ order ferromagnetically with Curie temperatures around 100 K and saturation magnetization of ~ 0.2 emu/g. Although the first report of ferromagnetism in this system utilized samples prepared by annealing an amorphous precursor, Srinivas and Dunlap⁶ subsequently showed that ferromagnetic quasicrystals in this system could be prepared by direct quenching from the melt. The low field magnetization of a typical ferromagnetic Al-Mn-Si alloy is illustrated in Fig. 1. These results show a behavior which is not well described by conventional models of ferromagnetism: $T^{3/2}$ behavior or a Brillouin function (see Fig. 1). The complexity of the ferromagnetism in these alloys is manifested in the high field magnetization as illustrated in Fig. 2. The paramagnetic Curie-Weiss behavior seen here at low temperatures is characteristic of ferromagnetic Al-Mn-Si quasicrystals.

The effects of other (non-Mn) transition-metal substitutions in quasicrystals have been considered extensively for paramagnetic alloys (see, for example, Refs. 7 and 8). It has been shown in these paramagnetic Al-Mn-based quasicrystals, that small quantities of Fe typically substitute for non-magnetic Mn and do not themselves carry a magnetic moment. As illustrated in Fig. 3, we see that the presence of Fe in ferromagnetic quasicrystals significantly affects the magnetization curve. A comparison with Fig. 1 shows that the addition of 2 at. % Fe has (i) lowered T_c , (ii) lowered the saturation magnetization, and (iii) enhanced the low-tem-

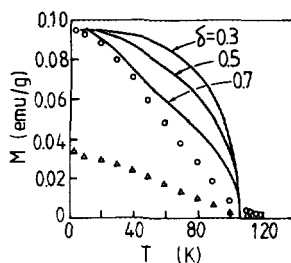


FIG. 1. Magnetization of icosahedral $\text{Al}_{50}\text{Mn}_{20}\text{Si}_{30}$ measured in applied fields of 20 Oe (Δ) and 50 Oe (\circ). Solid lines are computed data of Brillouin function corresponding to the 50-Oe data for $S = \frac{1}{2}$ and different values of δ , where δ is the rms exchange fluctuation.

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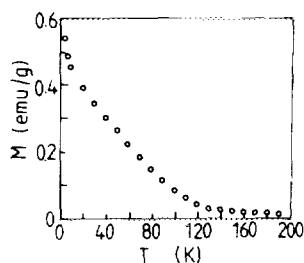


FIG. 2. Magnetization of icosahedral $\text{Al}_{40}\text{Mn}_{20}\text{Si}_{40}$ measured in an applied field of 10^4 Oe.

perature (nonferromagnetic) component of the magnetization. In this case the magnetization shows a low-temperature cusp at about 12 K characteristic of spin-glass ordering. Another example of an Fe-substituted Al-Mn-Si ferromagnetic quasicrystal is shown in Fig. 4. In this case the paramagnetic/spin-glass behavior is dominant and the ferromagnetism merely causes a slight inflection of the $M(T)$ curve at around 60 K.

B. Al-Mn-Ge-based alloys

A ferromagnetic alloy of the composition $\text{Al}_{40}\text{Mn}_{25}\text{Cu}_{10}\text{Ge}_{25}$ was first reported by Tsai *et al.*² This and related alloys show T_c values around 400–500 K and magnetizations which are significantly larger than those seen in Al-Mn-Si quasicrystals. We have looked at the influence of Fe substitution in the $\text{Al}_{40}\text{Mn}_{25}\text{Cu}_{10-x}\text{Fe}_x\text{Ge}_{25}$ system.

The saturation magnetization of these alloys, as illustrated in Fig. 5, is found to be a sensitive function of Fe content. In all cases, these alloys show a spin-glass component at low temperatures superimposed on the ferromagnetism.

A typical ^{57}Fe Mössbauer-effect spectrum of one of the alloys in the ferromagnetic region is illustrated in Fig. 6. A hyperfine field distribution, as seen in the figure, has been extracted from the spectrum by expanding probability distribution of hyperfine fields, $P(H)$, as a set of discrete points. A mean hyperfine field of 17.5 kOe has been obtained by averaging $P(H)$. In-field Mössbauer measurement in the $x = 3$ alloy showed the sign of the field to be positive. In contrast to the magnetization the mean Fe hyperfine field is found to be essentially independent of the Fe content of these alloys.

IV. DISCUSSION

Two major questions can be posed concerning the magnetic properties of ferromagnetic quasicrystals as reported here: (i) what is the origin of the ferromagnetism, and (ii) how is the additional magnetic component at low temperatures related to the ferromagnetism. The early reports of

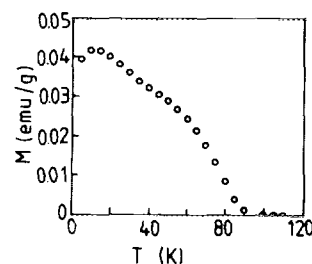


FIG. 3. Magnetization of icosahedral $\text{Al}_{48}\text{Mn}_{20}\text{Fe}_2\text{Si}_{10}$ measured in an applied field of 100 Oe.

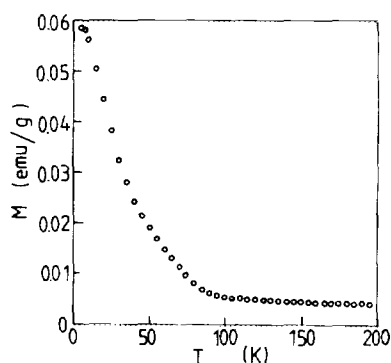


FIG. 4. Magnetization of icosahedral $\text{Al}_{42}\text{Mn}_{25}\text{Fe}_3\text{Si}_{10}$ measured in an applied field of 100 Oe.

ferromagnetic order with saturation magnetization of ~ 0.2 emu/g (Al-Mn-Si)³ or even ~ 2 emu/g (Al-Mn-Cu-Ge)² generated some concern that the ferromagnetism could be due to an impurity phase; this would explain the high values of T_c (with corresponding small M_s) and the relative insensitivity of T_c to composition. Evidence against this possibility includes, (i) lack of impurity peaks in x-ray diffraction studies, and (ii) lack of compositional inhomogeneities at grain boundaries in electron microscopy studies.⁹ With new Al-Mn-Cu-Fe-Ge quasicrystals with M_s approaching 20 emu/g, the possibility that this is due to an impurity phase seems to be ruled out. The ferromagnetism must therefore be considered to be an intrinsic property of the quasicrystalline phase.

The paramagnetic/spin-glass component seems to be characteristic of all ferromagnetic quasicrystals reported thus far. Although this in itself is not confirming evidence that the nonferromagnetic component of the magnetization is intrinsically related to the magnetic ordering, ferromagnetic resonance studies have helped to elucidate this problem in Al-Mn-Si quasicrystals. Misra *et al.*⁵ have reported the temperature dependence of a magnetization deduced from the effective g factor of a single resonance line. This shows the characteristic upturn at low temperatures as seen for example, in Fig. 2. As a single resonance line should represent the behavior of a single species of spins it would seem, therefore,

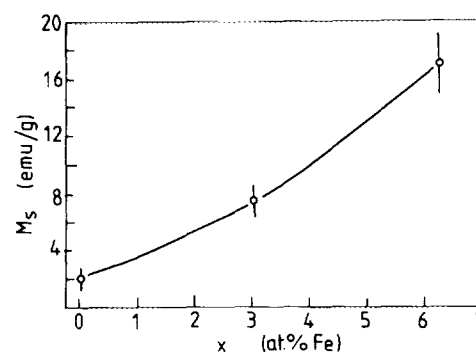


FIG. 5. Fe dependence of the 5×10^4 Oe magnetization for icosahedral $\text{Al}_{40}\text{Mn}_{25}\text{Cu}_{10-x}\text{Fe}_x\text{Ge}_{25}$. Data for $x = 0$ is extrapolated from the curve reported by Tsai *et al.* (Ref. 2).

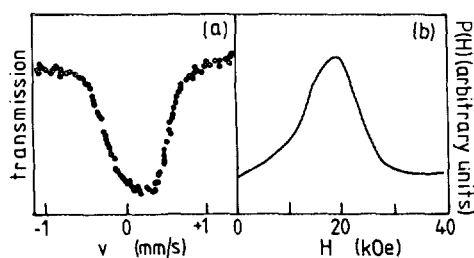


FIG. 6. (a) Room-temperature ^{57}Fe Mössbauer-effect spectrum of icosahedral $\text{Al}_{40}\text{Mn}_{25}\text{Cu}_4\text{Fe}_6\text{Ge}_{25}$. (b) Fe hyperfine field distribution.

that the ferromagnetism and the anomalous low-temperature magnetization were characteristic of the same magnetic phase. Future studies (for example, neutron diffraction) will hopefully provide clues as to the nature of this novel behavior.

Present studies of Fe-substituted alloys, have shown that the bulk magnetic properties are sensitive to composition. On the other hand, internal Fe hyperfine fields do not seem to vary appreciably with Fe content of the alloy. These observations are, perhaps, difficult to reconcile. Ongoing

studies of Fe hyperfine field systematics in alloys with a variety of compositions and a comparison of these to models for local and transferred hyperfine field contributions in ferromagnetic materials (e.g., RKKY interactions) will, we believe, help to clarify this behavior.

ACKNOWLEDGMENTS

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