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Measurements of Pressure Dependence of Magnetic Moment in Iron–Aluminum Alloys

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A method of measuring the change in magnetic moment with pressure for spherical samples at a relatively low pressure level is described. Special attention is paid to the problem of avoiding the effects of sample motion with respect to the detector coil to insure the accuracy in such a highly sensitive measurement. The results of measurements on iron–aluminum alloys up to 30 at.% Al are presented here. There is a sharp increase in the pressure dependence of the magnetic moment with the aluminum concentration. This is interpreted as favoring a competing antiferromagnetic interaction with increasing pressure.

In a model[1] which qualitatively accounts for the complex magnetic behavior of the iron–aluminum alloys as a function of temperature and composition, two competing exchange interactions are considered. These are a ferromagnetic interaction between nearest-neighbor iron atoms and an antiferromagnetic one between fourth-neighbor (across-the-body diagonal) iron atoms wherever an Al atom intervenes. When the alloys order, the Fe atoms occupy the cube corners and the cube centers are populated by Al and Fe atoms. In the limit of FeAl the only interaction in the model is the antiferromagnetic one between the Fe atoms on the cube corners through the aluminum atoms on the center sites. For smaller Al concentrations the ferromagnetic interaction depends on the presence of the iron atoms in the cube centers. The antiferromagnetism may arise from a superexchange via the $3p$ wavefunctions of the Al. The ferromagnetism is thought to occur when $3d$ bands form because of the overlap of wavefunctions from nearest-neighbor iron atoms. That is, in the limit of FeAl the smaller overlap of wavefunctions from the second-neighbor iron atoms results in $3d$ bands which are presumed not to favor ferromagnetism. One may therefore expect that in the Fe(Al) series a decrease in volume would primarily lead to an increase in the antiferromagnetic superexchange interaction. The model has some resemblance to the picture used to discuss the volume dependence of the magnetic transition points of Eu-chalcogenides[2] in the sense a two competing interaction model is adopted, although the roles of the two effects are reversed in the latter.

The Fe(Al) alloys are ferromagnetic at low temperatures for less than ~28 at.% Al and antiferromagnetic for more than ~32 at.% Al. In the transition region they appear antiferromagnetic at low temperatures, ferromagnetic near room temperatures, and paramagnetic at high temperatures.[3] The data reported here are for alloys in the ferromagnetic region.

METHOD OF MEASUREMENT

A uniformly magnetized sphere produces an external field identical to that of a dipole of total moment $\mu_T = m\sigma = MV$. The inductive coupling of this moment to a detector coil depends only on the position of the center of the sphere with respect to the coil. An apparatus calibrated for one sphere is calibrated for all spheres. If an externally applied field is sufficient to saturate the magnetization, the application of pressure will produce a change in total moment $\Delta\mu_T = m(d\sigma/dP)\Delta P$. But below saturation, if the permeability is high, M depends only on the applied field, B_a, with the result that $\Delta\mu_T = (3B_a/4\pi)(dV/dP)\Delta P$. One can determine magnetically both the compressibility and the pressure dependence of the saturation magnetization.

The methods discussed here of measuring the change of μ_T with pressure were developed in order to study $d^2\sigma_s/dPdc$, where c is the concentration of various solutes in Fe.[4] The stringent requirements of that work made clear several difficulties. It is most important to minimize effects from changes in the position of the center of the sphere with pressure. As we measure directly the integral of the induced voltage in the detector coil during the application of pressure, it is necessary to integrate over a time long compared to that for the sample to return to its initial temperature, that is for several minutes. (The adiabatic change is 50% larger than the isothermal change in the case of pure Fe.)

The detector coil is made in two matched sections which can be connected in either series opposition or series aiding. If the sections are in series opposition, the sample is positioned by requiring that no signal results from changes in sample magnetization when an applied field is swept at constant rate. A small voltage arises from the fact that the coils are not perfectly matched. But this is independent of the sample magnetization which changes rapidly at low fields and little at high fields, so that it is sufficient to adjust the position of the coil with respect to the sample until this small voltage depends only on the rate of change of the applied field and not on the field itself. After the sample is magnetically centered with respect to the sections of the coil, the sections are connected in series aiding and one obtains maximum sensitivity to changes in μ_T and minimum sensitivity to small changes in the position of the sphere. We use two such identical sets of coils with matched sections. One contains a reference sample of Fe, the other holds the alloy sample. These are connected in series opposition so that no signal results from the application of pressure when there are iron samples in both coils subjected to the same changes in pressure. While this method of positioning the coils works well in almost every case, there was one ball in which the magnetic center changed with field, presumably because the domain structure was not sufficiently homogeneous.

THE APPARATUS

Berylium copper sample holders are pressurized to 0.5 kbar by pneumatically pumped oil. The detector coils have 3000 turns in each section of an aluminum coil form. Each section has an i.d. of $\frac{9}{16}$ in., an o.d. of $\frac{3}{4}$-in. and a width of $\frac{1}{8}$ in. The signal from the detector coils is amplified using a Kiethley 147 nanovoltmeter and integrated using an Analogue operational amplifier. The integrated voltage shows a rapid change as the pressure is applied followed by a slow relaxation to a smaller value during the return to the sample temperature to its initial point. The return is assured by using a large thermal mass. There is usually a slight change with pressure in the dc level in the input circuit which shows up as a change in output voltage which is linear in time after the pressure is turned on. Therefore, it is necessary to extrapolate a linear change in the integrated voltage back to the time that the pressure was applied. The apparatus is calibrated by measuring the signal induced when the magnetization of the sphere changes with applied field.

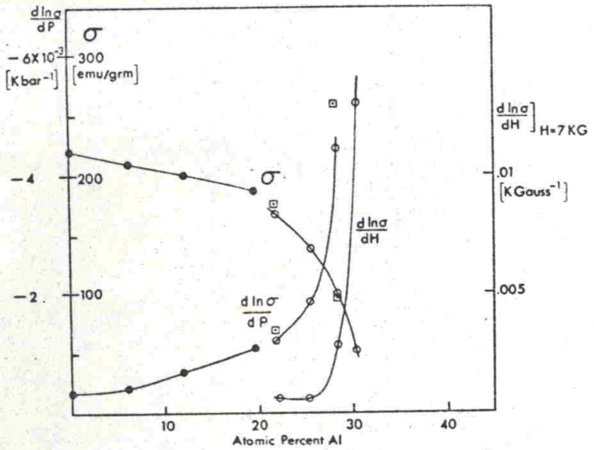

FIG. 1. The variation of saturation magnetization and its dependence upon pressure and upon applied field for iron-aluminum alloys measured at room temperature. ○: slowly cooled disordered alloys; ●: slowly cooled alloys ([Fe_3Al] ordered structure); □: quenched alloys ([FeAl] ordered structure).

RESULTS

Figure 1 shows $-d\ln\sigma/dP$ for the ferromagnetic iron-aluminum alloys at room temperature. Measurements of the saturation magnetization[3] carried out on the same series of alloys (except for a 12-yr anneal at room temperature) are added for reference. The alloys below 20% are well annealed but in the disordered phase. The alloys above 20% are well annealed and very slowly cooled to produce the [Fe_3Al] phase. Two of the specimens were well annealed and then quenched. This produced the [FeAl] ordered phase for the 28.3% sample, but the degree of order of the 22% sample is not certain.

The rapid rise in $-d\ln\sigma/dP$ is not just due to the drop in σ, for $-d\sigma/dP$ itself shows a sharp increase at 28.3%. This increase correlates with the increase in the susceptibility above saturation[3] (also shown in Fig. 1). This correlation thus indicates that, in the delicate balance between the ferromagnetic and antiferromagnetic interactions used to explain the complicated magnetic behavior of these alloys, the applied field favors ferromagnetism[1] while the volume decrease from pressure favors antiferromagnetism. One cannot conclude with the present experiment alone whether this change is due to the effect of pressure on the saturation magnetization at absolute zero or to the effect on the Curie point inasmuch as the law of corresponding states does not apply for these higher aluminum content alloys. It should therefore be informative to carry out the pressure measurements as a function of temperature for an alloy in the transition range of composition.

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