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PROGRESS REPORT
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THE RELATIONSHIP BETWEEN MICROSTRUCTURE AND MAGNETIC PROPERTIES IN HIGH-ENERGY PERMANENT MAGNETS CHARACTERIZED BY POLYTWINNED STRUCTURES

Submitted by:

Professor William A. Soffa
Department of Materials Science and Engineering
University of Pittsburgh
Pittsburgh, PA 15261

First Report of (Second) 3 Year Grant
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Attention: **Dr. Michael E. Kassner**
Division of Materials Science
Office of Basic Energy Sciences

PROGRESS DURING THE PAST YEAR

During the past year the research program has taken on some new and exciting dimensions. The program has been directed at understanding the relationship between the polytwinned microstructures which develop during ordering ($A1 \rightarrow L1_o$) in the Fe-Pd and Fe-Pt ferromagnets and their technical magnetic properties. The research has revealed for the first time the salient features of the magnetic domain structures which emerge within the polytwinned state and has analyzed the mechanism of coercivity in terms of a modified APB pinning model including thermal activation. The consideration of thermal activation explains for the first time the steep temperature dependence of the coercivity [1]. However, recently T. Klemmer in his doctoral thesis work on the Fe-Pd alloys has shown that significantly enhanced coercivities are obtained in thermomechanically processed bulk and melt-spun alloys which exhibit recrystallized grain sizes $\leq 1\ \mu m$ and do not contain the so-called c-domains (transformation twins). The fine-grained materials show coercivities of about double those generally reported for bulk processed materials and clearly these are not optimized properties. Actually, a careful re-examination of the literature shows that a few papers in the Russian literature reported the development of enhanced coercivities in fine-grained Fe-Pd alloys and in one study even called attention to the absence of APB's and transformation twins in recrystallized grains [2-4]. Also, coercivities approaching 2kOe have been reported in ultra-fine grained Fe-Pd thin films [5,6]. These data were buried in the literature mainly because they stemmed from preliminary studies of a rather perfunctory nature and there was no systematic, in-depth, follow-up to the work. In particular, virtually no microstructural studies were pursued to elucidate the structure-properties basis for these properties nor was there any meaningful discussion of the change in the mechanism of coercivity which apparently had occurred. Importantly, in these Russian reports and in the current studies the change in the microstructure and properties indicate a change in the mechanism of coercivity when the grain diameter of these polycrystalline ferromagnets reaches D ~ 1 μm, as mentioned above. Thus, it is interesting to estimate the critical diameter, D_c, the so-called "single-domain particle diameter", for the $L1_0$ ferromagnets. Grain sizes $D > D_c$ are expected to be multi-domain, whereas when $D < D_c$ each grain in the polycrystalline aggregate is likely to be a single magnetic domain and not contain magnetic domain walls. A good estimate of the critical dimension, D_c, for high anisotropy materials is $D_c = \frac{2K\delta}{\pi^2 M_s^2}$, where K is the crystal anisotropy constant, δ is the domain wall thickness, and M_s is the saturation magnetization. Taking values for the Fe-Pd alloy, $K \sim 3 \times 10^7$ ergs/cm^3, $\delta \sim 6 \times 10^{-7}$ cm, and $M_s \sim 1000$ emu/cm^3 gives the result $D_c \sim 0.1$ μm. This calculation is for an isolated cube of edge D. For a uniaxially magnetized sphere a similar calculation yields a critical diameter $D_c \sim 0.5$ μm. Such calculations are essentially uncertain within a factor of about 2. Thus, fine-grained polycrystalline aggregates of these high anisotropy materials with grain diameters $D \lesssim 1$ μm are approaching single-domain behavior and new mechanisms of coercivity are expected to control the magnetic hysteresis behavior. Since the small grains are found to contain no c-domain structure (structural domains or transformation twins) and only exhibit coarse {111} annealing twins, even if the grains support a simple domain structure the pinning mechanism is likely to be very different compared to the polytwinned state.

Figures 1 and 2 show recrystallized grains of the equiatomic Fe-Pd alloy cold-rolled approximately 80% in the disordered state and aged for 100 minutes at 500°C. The recrystallized grains clearly do not show the polytwinned structure and exhibit only coarse annealing twins. Importantly, the annealing treatment at 500°C occurs at a temperature where the ordering and recrystallization processes occur concomitantly.

The recrystallization of the $L1_0$ ordered ferromagnets to produce ultra-fine grain sizes and enhanced magnetic properties has important implications far beyond the limited context of these unique high anisotropy permanent magnet materials. The renewed focus on ordered phases, in general, such as Ni_3Al, NiAl, Fe_3Al, Ti_3Al and TiAl for structural applications at high temperatures has rekindled interest in the physical metallurgy of orderable alloys and intermetallic compounds. During processing and/or in service, the recovery, recrystallization, and grain growth behavior of these materials is of primary interest. However, it is clear that there are complex interactions between ordering and the structural relaxation processes associated with annealing in both "reversibly" ordered and "permanently" ordered intermetallic phases [7]. A number of older studies of recrystallization in ordering alloys clearly indicated that coldworked materials exhibited retarded recrystallization kinetics when ordering of the deformed matrix occurs prior to recrystallization. Some investigators suggested that ordering completely suppresses recrystallization below the critical ordering temperature, T_c, of the particular alloy system. Generally, what is occurring is that ordering markedly reduces grain boundary mobility by a factor of a hundred-fold or more as well as influencing the dynamics of dislocation rearrangement required to produce recrystallization "nuclei" which can grow spontaneously and consume the coldworked state [8,9]. The results of the past year on the coldworked and recrystallized $L1_0$ ferromagnets in this work clearly suggest that the synergistics of the combined ordering and recrystallization processes can be used to produce ultra-fine grained materials with extraordinary properties.

Finally, the microcrystalline, high coercivity state which develops in the ordered $L1_0$ ferromagnets mimics the isotropic, microcrystalline-type structures produced by melt-spinning rare earth permanent magnets. Each grain in these aggregates is a single domain but magnetization reversal may occur by the motion of 180° walls across each grain. Virtual "domain walls" essentially exist at each grain boundary and emerge into the grains under the influence of the applied field. Each grain may reverse independently or along an "interaction domain" boundary. This mechanism of coercivity may be important in a number of magnetic materials but has not been investigated comprehensively either experimentally and theoretically.

PUBLICATIONS

"Magnetic Domains and Coercivity in Polytwinned Ferromagnets", B. Zhang and W.A. Soffa, Phys. Stat. Sol. (a), 131, 707 (1992).

"The Structure and Properties of $L1_0$ Ordered Ferromagnets: Co-Pt, Fe-Pt, Fe-Pd and MnAl", B. Zhang and W.A. Soffa, submitted to Scripta Met., special issue on Magnetic Materials (in press).

INVITED TALKS

TWINNING IN ADVANCED MATERIALS, Symposium at 1993 TMS Fall Meeting

"Magnetic Domains and Magnetization Reversal in Polytwinned Ferromagnets"

PROJECT EMPHASIS DURING THE NEXT YEAR

A primary thrust of the next grant period will be to focus on the influence of the grain size on the magnetic properties of the $L1_0$ ordered ferromagnets when the grain diameters approach 1 μm and less. The ultra-fine grained structures will be produced generally by coldworking and annealing both bulk and melt-spun alloys utilizing the synergistics of these solid state transformations. Also, ultra-fine grained thin films will be produced by vacuum sputtering. Thus, the project now will include a new component, that is, the systematic study of the influence of ordering on the fundamental processes involved in the recrystallization of these orderable alloys and how the combined reactions of ordering and recrystallization can be exploited to tailor the structure and properties of these magnetic materials.

Also, the forthcoming year will involve the development of models to describe the mechanisms of coercivity which are likely to control magnetization reversal and hysteresis in the regime of fine grain size. This new facet of the structure-property studies will include Lorentz microscopy studies of the changes in magnetic domain structure which accompany the disappearance of the c-domains characteristic of the polytwinned state. As mentioned above, particular attention may be directed at magnetization reversal possibly effected by the movement of "interaction domain" boundaries.

Importantly, work will continue on the polytwinned state. The Lorentz microscopy observations of the unique magnetic domain structures which emerge in polysynthetically twinned ferromagnetic crystals will continue including more work on the Fe-Pt alloys. Thin foil preparation for transmission electron microscopy has been somewhat problematic with the Fe-Pt system but that appears to have been overcome. Finally, studies of the temperature dependence of the coercivity in different regimes of hysteresis behavior and the nature of the elementary thermally activated processes will be extended.

PERSONNEL

Three full-time graduate students will be affiliated with the program over the next grant period. Below is a brief synopsis of their academic backgrounds and status.

Mr. Timothy J. Klemmer

Mr. Klemmer is a 1989 graduate of the Department of Materials Science and Engineering here at the University of Pittsburgh. He has passed all of his course work and qualifying examinations for the Ph.D. degree and is working full-time on the project for his doctoral thesis.

Mr. David Hoydick

Mr. Hoydick is a 1991 Honors Graduate of the University of Pittsburgh majoring in Engineering Physics. He is working on the project for his doctoral thesis in Materials Science.

Mr. H. Okumura

Mr. Okumura completed his Master's degree in Materials Science at Kyoto University in Japan in 1989 and working on the project for his doctoral thesis research.

LIST OF FIGURES

Figure 1

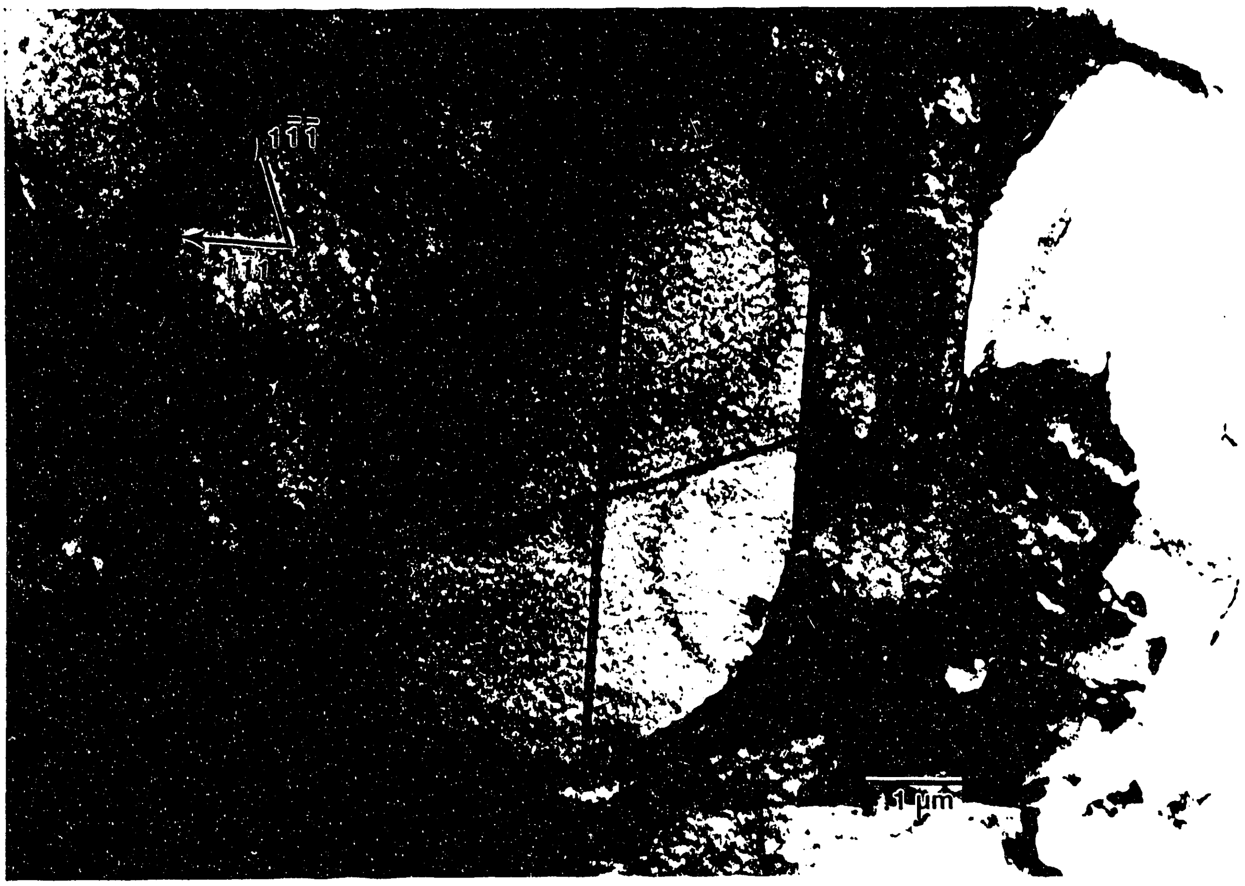

Figure 2

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