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On

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Rare Earth-Iron Materials.

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INTRODUCTION

Personnel

During the past contract year the personnel associated with the program have changed to some extent, and consequently the program has shifted somewhat in emphasis. William Pala, whose dissertation under Professor Romeo Segnan was on the magnetic structure of amorphous YFe$_2$, through Mössbauer studies, finished, received his Ph.D., and moved on. We are pleased to believe that Dr. Pala’s work revealed something significant – that amorphous YFe$_2$ is a spin glass. Recent studies, by others, on YFe$_x$ show that below a threshold iron concentration there is no moment on the iron, while at larger $x$, encompassing YFe$_2$, the iron has a moment, but one frozen in a disordered spin glass state.

Dr. Y.J. Liu, who was at one time working full time with us on amorphous materials and magnetoelastic surface waves on conducting surfaces, now works at the Naval Surface Weapons Center on the cross tie memory, under other funds. His place in our program has been occupied for the past year by Dr. Stefano Rinaldi of the MASPEC Laboratory, Parma, Italy.

Another person who has been working with us (on a limited basis because of the shortage of funds), is Dr. Kristal Hathaway. Dr. Hathaway studied with Professor Krumhansl at Cornell. For us, she has been calculating phonon spectra in Cl5 structure compounds (cubic, Laves phase, the RFe$_2$ structure), and introducing magnetoelastic coupling to learn what happens to the spin wave and phonon branches when the coupling is strong, as it is in these materials.

Program

Dr. Rinaldi joined us just as the RFe$_2$ elastic properties research “got hot,” and he has spent most of his year on ultrasonics measurements. This has been productive, as the attached publications will show. We have observed something not seen previously – symmetry breaking by the magnetization in a disordered system.
Suppose the magnetization to lie along the \( z \) axis and the propagation direction of a shear wave to be the \( y \) axis. Because of magnetoelastic coupling symmetry is broken and there are two distinct shear modes, one polarized along \( x \), and one along the magnetization axis, \( z \).

In the \( \text{RF}_{2} \)'s, with their enormous magnetostriction, the velocity of sound for the two modes can differ by 50%. Dr. Rinaldi has also observed that application of a magnetic field can change the velocity of sound by 50% and more. This would of course be the result if one used the field to rotate the magnetization with respect to the phonon polarization, given the two modes of different velocities. But there is also another effect, for fixed field direction. As the phonon passes down the crystal, the local easy axis rotates back and forth. This is because of the distortion, the magnetostriction, and the \( K \) effect. Since the phonon passes slowly compared to the Landau-Lifshitz relaxation time, the spins point along their local easy axis, following the phonon, and waving back and forth like wheat in a wind storm. Rotation of the spins has a second order feed-back on the effective elastic constant. If one pins the spin in a fixed direction by a magnetic field, the elastic constant is larger - over 50% larger in our materials - than if one allows the spins to relax to their lowest energy direction. Through field modulation, one hereby has a tuneable acoustic. There is also a thought of other applications, such as a switch.

In some ways (huge magnetoelastic coupling) the \( \text{RF}_{2} \) materials are superior as magnetostrictive transducer materials to other conventional magnetic substances, such as the garnets. But in other ways (magnetic anisotropy, electrical conductivity) they are much worse.

The deleterious effect of high magnetic anisotropy is being reduced by compounding rare earths of the same sign of \( \psi \) but opposite signs of \( K \). Thus, by judicious mixing one obtains a hitherto-unapproached magnetomechanical coupling figure of merit \( \left( \frac{\psi}{K} \right) \) of 0.66, for \( \text{Tb}_{2}\text{Dy}_{0.22}\text{Ho}_{0.58}\text{Fe}_{2} \).
The electrical conductivity, and consequent skin depth and eddy current loss problem remains. This was of concern to Dr. Y.J. Liu, who investigated circumstances under which losses would be tolerable for surface wave devices. A question we hope to address in the following year is optimum design of composites — RFe₂ particles embedded in a non-conducting matrix. How much RFe₂? What shape particles? What elastic and magnetic properties does one desire of the matrix? This is a difficult (in fact, insoluble) problem. It is reminiscent of the question of the elastic properties of polycrystals, a problem only approximately solved in limiting cases. But in designing composites or in deciding on the utility of the whole concept, even qualitative insights would be helpful.

Another area we have explored is the magnetic properties of the random anisotropy model. Our molecular field theory results are in contradiction with the computer calculations of Harris and Zobin,¹ of Chi and Alben,² and now of Patterson, Gruzalski and Sellinger (Phys. Rev. Letters, to appear), all of whom also agree and disagree with each other.

First, as to the magnetic ground state: Harris and Zobin, alone of the four, find a spin glass state lying lowest, above a threshold ratio of anisotropy to exchange. This seems unlikely to us, but cannot be dismissed out of hand. The other three papers find a magnetically ordered state to be that of lowest energy. The remanence depends upon D/J, and since the computer calculations have been performed for different values of parameters, comparison is not possible. But there are clear differences. We find the remanence decreasing monotonically to 0.5 as D/J increases. Chi and Alben find the remanence to drop below 0.5 and then to approach 0.5 asymptotically as D/J rises. Patterson et al., are silent on the general functional dependence of remanence on D/J, but at the particular value of D/J for which they evaluate the remanence, they find it to be greater than at its infinite D limit, the
Iropic upper hemisphere average. And Harris and Zobin find the remanence to drop discontinuously to zero at a particular D/J, at the transition to a spin-glass state.

There is also discord about the coercive force. Harris and Zobin calculate the initial magnetization and characterize a jump in that quantity as the coercive force. Chi and Alben find the coercive force to rise abruptly and saturate at a low value, independent of D/J, for all D/J after the rise. We find (and we consider this to be erroneous, an artifact of molecular field theory) there to be a threshold D/J below which the coercive force is zero. Molecular field theory, a single particle theory, misses the collective behavior. If all the spins rotated rigidly as they almost do at small D, there would be no coercive force. All directions of M would see the same uniform distribution of easy axes. At the other extreme, where D/J is very large, however, we find the coercive force, at T=0°K and ignoring relaxation effects, to rise linearly with D. This should surely be correct when all the important terms are single ion terms and the coercive force is merely the energy to lift enough spins over their anisotropy barriers, from metastable states to their lowest energy states along the field. Inexplicably (to us) the saturation claimed by Chi and Alben, of coercive force, is confirmed by Patterson, et. al., in a calculation based on equilibrium statistics. How can a coercive force be found in equilibrium? In equilibrium the moment must lie along the field. (Of course the introduction of relaxation times and of domain nucleation effects will change all of this.) There is work to do on coercive force on the random anisotropy model, and much work on thin films, on permalloy, on magnetization ripple to be rediscovered by a new generation. But enough on random anisotropy.

In the RFe₂ amorphous materials, not all the disorder comes from the randomly oriented anisotropy axes. There is also exchange disorder. We have shown that
the magnetization and Curie temperature data on Tb$_x$Fe can be explained by considering the amorphous Fe to have an average z component of moment of 0.8 $\mu_B$ (although each Fe has moment of magnitude 2.2 $\mu_B$; there are some negative exchange interactions which cause the iron moments to lie off at various angles from the average direction). Each Tb ion added, through its antiferromagnetic interaction with its neighboring irons, causes the cluster of surrounding irons to straighten up (or rather down - the iron direction is taken to be down), thus reducing the effect on the net moment of adding a Tb, with its 9 $\mu_B$ moment, in the "up" direction. This yields the observed moment dependence of the concentration if one takes the number of iron neighbors of a rare earth in the amorphous phase to be 6.

But is this the correct number? Cargill's x-ray studies suggested it to be so, and it is plausible on geometric packing grounds, but we felt it worthwhile to get evidence independent and more direct than the x-ray or neutron data. Hence work was performed at SLAC with Professor Edward Stern, of the University of Washington, on EXAFS studies of a number of amorphous RFe$_2$ samples. A preliminary report of this research was given, at the International Conference on Magnetic Alloys and Oxides, Haifa, August, 1977. (6 is about right.)

One interesting conclusion, yet to be pursued, is that the phase shift in the amorphous material is distinctly different from that in the crystal. Some of the distances reported in x-ray studies tentatively seem to be not quite right, also.

Another problem which appears to be worth some effort is the analysis of the magnetoelastic properties of strongly coupled systems from a non-perturbative viewpoint. Usually, one has weak coupling, and treats this as a perturbation. But in the RFe$_2$'s this is unjustified. This is a soluble problem, but messy. One must diagonalize a quadratic form. This leads to a different figure of merit than the usual $\chi^2/\kappa$, which of course is not right when one dopes the material so that its average $\kappa$ is zero. This is well known, but there are aspects still to be
explored, at large $\lambda$.

This is but a skimpy review of what has been done in the past year, because those efforts have mostly been reported in the literature. The next sections list those of our publications during the past year supported by ONR, and over the earlier years of the contract. We will gladly send copies of publications to those who request them.

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1. R. Harris and D. Zobin, AIP Conf. Proc. 29, 156.

Publications Supported by ONR During 1976-77


2) K. Sato, E. Callen and N. Tamagawa, Magnetic Properties of (Fe-Co)-Tb Alloys, J. College of Liberal Arts, Toyama University, Japan, 2, 19 (1976).


Publications Supported by ONR Contract From Inception to 1976


