

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

AD-A244 422



tion is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, preparing and reviewing the collection of information, sending comments regarding this burden estimate or any other aspect of this reducing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503

2. REPORT DATE Nov. 22, 1991	3. REPORT TYPE AND DATES COVERED Final Report June 88 - Sept. 91
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Magnetic Structures and Excitations in Thin Films and Multilayers	5. FUNDING NUMBERS DAAL03-88-K-0061 ②
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8. PERFORMING ORGANIZATION REPORT NUMBER
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9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P. O. Box 12211 Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER ARO 25374.14-PH

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.
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12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)
Summary of work performed during 3 year period. Main topics include
1) Gigantic magnetoresistance in magnetic multilayers
2) Properties of ultra-thin magnetic films
3) Infra-red properties of magnetic materials and doped semiconductors
4) Phase transitions and the tailoring of magnetic properties in magnetic multilayers.

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JAN 09 1992
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14. SUBJECT TERMS Magnetic multilayers, ferromagnets, antiferromagnets, magnetoresistance, infra-red, monolayers, magnetization
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15. NUMBER OF PAGES 10

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED

18. SECURITY CLASSIFICATION UNCLASSIFIED

19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED

20. LIMITATION OF ABSTRACT UL

Magnetic Structures and Excitations in Thin Films and Multilayers

Final Report

by

R. E. Camley

November 22, 1991



U.S. Army Research Office

Contract # DAALO3-88-K-0061

University of Colorado
at Colorado Springs

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DTIC - TAB	<input type="checkbox"/>
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92-00440



92 1 7 074

Statement of the Problems Studied

This project involved a general theoretical study of the properties of magnetic multilayer systems. Four major areas can be identified:

- 1) Gigantic magnetoresistance in magnetic multilayered systems
- 2) Properties of ultra-thin (few atomic layers) magnetic films
- 3) Electromagnetic properties of magnetic and semiconductor multilayers in the infra-red.
- 4) Tailoring the magnetic properties of magnetic multilayers

Below we discuss each of the four areas in more detail.

Gigantic magnetoresistance in magnetic multilayered systems

In 1989 it was discovered that in Fe/Cr multilayer structures a surprising effect occurs. In sandwich structures of $100 \text{ \AA} \text{ Fe} / 10 \text{ \AA} \text{ Cr} / 100 \text{ \AA} \text{ Fe}$ a change in resistivity with field of up to 3% was observed. **In Fe/Cr multilayer structures an astonishing magnetoresistivity effect of up to 50% was measured at low temperatures.** These numbers should be compared with a magnetoresistivity effect of about .2% for thin Fe films. In the Fe/Cr multilayers, there is an effective antiferromagnetic coupling of the Fe films due to the intervening Cr films. As a result, the magnetic moments of neighboring Fe films are antiparallel to each other in zero field, but may be forced to lie parallel in a strong field. The resistance of the multilayer is largest when the magnetic moments in neighboring Fe films are antiparallel and smallest when they are parallel. Since the earliest reports a lot of experimental work has been carried out and other systems with a magnetoresistivity effect up to 60% even at room temperature have now been discovered. This effect has significant potential for use as sensors and in magnetic memories. In this portion of the work we developed a theoretical basis by which this new effect could be understood.

Properties of ultra-thin (few atomic layers) magnetic films

A number of basic issues were studied for ultra-thin films. For example, the origin of long range magnetic order in ultra-thin films was explored. A monolayer of spins coupled by Heisenberg exchange interactions only should never exhibit long range order at finite temperatures according to fundamental principles of statistical mechanics. Yet experimentally monolayers of magnetic ions on metallic substrates are found to order, even at room temperature. A second issue is the nature of the ground state and spin excitations (spin waves) in ultra-thin films. If we consider a monolayer, with easy axis anisotropy normal to the surface, then in the ordered state the magnetization is normal to the surface. As layers are added (say by 4 - 5 layers) the magnetization has rotated to be parallel to the surface as a consequence of a competition between anisotropy and dipolar interactions. Such behavior is known in thin films of Co for example. There is the possibility that at intermediate thicknesses, the magnetization is canted.

Electromagnetic properties of magnetic and semiconductor multilayers in the infra-red.

The spin wave excitations in antiferromagnets and plasma excitations in doped semiconductors have frequencies which places them in the infra-red range. In addition, magnetic systems (or magnetically controlled systems) have properties which make them particularly interesting for signal processing. First, since the frequency of the spin wave is generally dependent on the applied field, the response may be tuned by an external field. Second, magnetic excitations are often nonreciprocal and as such may be used in isolators and gyrators. In this portion of the work we studied infra-red reflection from semi-infinite antiferromagnets, antiferromagnets with rough surfaces, and doped semiconductor superlattices in a magnetic field. Particular attention was paid to nonreciprocal features.

Tailoring the magnetic properties of magnetic multilayers

Macroscopic properties of a material depend on the underlying microscopic ground state. In magnetic systems the fundamental properties of interest are the static susceptibility, the dynamic susceptibility, the magnetization as a function of temperature, and the compensation points (if any). All these material properties can be significantly adjusted by changing the layering pattern of a magnetic multilayer. In this portion of the work we studied systems (Fe/Gd and Gd/Dy) where significant changes in the macroscopic properties occur due to a **small** applied magnetic field. Direct contact to experimental measurements was made with excellent agreement between theory and experiment.

Summary of Most Important Results

Gigantic magnetoresistance in magnetic multilayered systems

In our theoretical work, we were able to explain the origin of the gigantic magnetoresistance in Fe/Cr multilayers as due to spin dependent diffusive scattering at the Fe/Cr interface. We computed the conductivity of the structure through the use of the Boltzmann transport equation. In addition to obtaining excellent magnetic field dependent results, we were also able to explain how the magnitude of the magnetoresistance effect depends on structure (it increases as the number of layers is increased) and on temperature (it increases by a factor of 2 to 3 as temperature is decreased from room temperature to liquid helium temperature). We later extended our earlier calculations to consider the possibility of spin-dependent bulk scattering as well as spin-dependent interface scattering. We were able to show that the fundamental contribution to the gigantic magnetoresistivity was indeed from the Fe/Cr interface scattering as previously assumed. We also extended our earlier calculations to include finite structures of the form $(\text{Fe/Cr})_n/\text{Fe}$ with n being the number of repeats of the Fe/Cr unit cell. Including both bulk and interface spin-dependent scattering and outer surface roughness scattering we are now able to obtain excellent agreement between theory and experiment.

Properties of ultra-thin (few atomic layers) magnetic films

We studied the question of the surprising magnetic thermal stability of ultra-thin films by two methods, a renormalization group method and classical Monte Carlo simulations. Both methods showed that anisotropy fields would stabilize the ferromagnetic state in even a monolayer thick film. We found that for reasonable anisotropy values the change in the transition temperature with thickness is quite rapid, with the ferromagnet reaching a transition temperature which is 90% of the bulk value by the time one has only 6 atomic layers. We also developed methods for linking the spin-wave equations of motion (for thin film and multilayers) to a transformation that casts the original Hamiltonian into diagonal form. Here we found some striking results. The interesting configuration is one where the film has an easy axis normal to the surface. In such a film application of a magnetic field parallel to the plane of the film will tilt that magnetization, and at a certain critical field the magnetization just touches the plane. At this critical field, we show long range order in the film must vanish, to be restored when the field is increased further in magnitude. This result is unique to ultra-thin films; for thick, quasi three dimensional films the singularity vanishes.

Electromagnetic properties of magnetic and semiconductor multilayers in the infra-red.

Here we studied reflectance of infra-red radiation from antiferromagnets and doped semiconductors. In the presence of a magnetic field both materials can exhibit nonreciprocal reflection. In the past, a thermodynamic argument had been made that absorption was necessary for nonreciprocal reflection to occur. This earlier calculation only considered the situation where the incident wave and the outgoing wave had the same polarization. By properly generalizing this argument to allow for mixing of polarizations we were able to show that absorption was not necessary for nonreciprocal reflection. We presented explicit calculations supporting this general argument. A number of specific examples were also studied. The major results from these examples include 1) surface roughness can enhance nonreciprocal effects by enhancing the coupling of the external waves to surface waves; 2) In the absence of absorption both circular and linear polarized light can be nonreciprocally reflected, but the frequency region where this occurs is quite narrow for linearly polarized light; 3) The reflectance from a semiconductor superlattice can change by a factor of 2 when the magnetic field is reversed.

Tailoring the magnetic properties of magnetic multilayers

Here we showed that the competition between Zeeman energy and exchange energy in magnetic multilayers could lead to a number of different microscopic magnetic states. One can "tune" to the different states by application of a small magnetic field. These states have different macroscopic properties such as static susceptibility and magnetization as a function of temperature, and we can predict (!) this behavior as a function of temperature, external field and layering pattern. To give some examples we quote from a recent experimental paper (Phys. Rev. B **44**, 7733 (91)) which studied an Fe/Gd system which we had treated theoretically much earlier (88-89). "The data presented here can obviously be interpreted on the basis of Camley et al calculations. The ...magnetic susceptibility... is maximum at $T_{\text{compensation}}$ and decreases as H_{critical} increases. This important result is in accordance with the theoretical calculations and is explained by the large orientational polarisability. In conclusion, it is manifest that the experimental results are in good agreement with theoretical predictions: i) the aligned and twisted states are present, ii) there is a compensation temperature, iii) the susceptibility is the largest at the onset of the twisted state, iv) the critical field for the aligned-twisted state transition goes close to zero at the compensation, v) H_{critical} increases when the thicknesses of the layers decreases."

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B. L. Johnson and R. E. Camley (submitted to MRS Symposium Proceedings)

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J. G. LePage	--	MS earned during project
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