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Correlation between Spin Polarization of Tunnel Currents from 3*d* Ferromagnets and Their Magnetic Moments

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We report measurements of spin-polarized tunnel currents from a wide compositional range of alloys of Ni with Fe, Mn, Cr, or Ti. The interpretation of the data indicates that all the alloys have an electron spin polarization P which is positive and whose magnitude follows closely the saturation magnetic moment of the alloy. This simple result greatly constrains possible explanations of spin-polarized tunneling from 3*d* ferromagnetic metals.

It was previously shown that the spin polarization P of the tunneling currents in elemental Ni, Co, and Fe is positive (predominantly in the majority spin direction).[1,2] Similar values of P for these metals had been obtained earlier from interpretations of the spin polarization of photoemitted electrons.[3] These results aroused great interest because they were apparently in direct contradiction to a simple interpretation of the band theory of ferromagnetism. This band-theory explanation would have the tunneling conductance for the two spin directions proportional to the magnitude of the two spin densities of states at the Fermi surface. For Ni the model predicts $P \approx -67\%$, which is wrong in sign as well as magnitude (measured P for Ni $\approx +10\%$). For Fe the magnitude of P in such a picture should be much less than for Ni, again contrary to experiment. Thus this simple band-theory explanation is incorrect. Because these unexplained results bear directly on the basic mechansim of ferromagnetism in the 3*d* metals, there has been much recent theoretical activity. Also experiments are in progress using tunneling, photoemission, field emission, and several other techniques. References to this current work are given in recent reviews.[4,5] Some preliminary results of Ni-Mn and Ni-Ti alloys were given in Ref. 5. The purpose of this Letter is to present measurements of the tunnel-current spin polarization over a wide range of 3*d* ferromagnetic metal alloys, which show that there is a striking correlation between the measured polarization and the saturation magnetic moment n_B of the alloys.

The technique and analysis of spin-polarized tunneling experiments have already been described in detail.[2] In essence the experiment consists of measuring the conductance dI/dV versus voltage V of a tunnel junction consisting of a 40-Å-thick superconducting Al film, an Al_2O_3 barrier, and the ferromagnetic metal film. With the junction in a parallel magnetic field of about 40 kOe and at a temperature of 0.4 K, the conductance shows an asymmetry of the spin-split tunneling conductance about $V = 0$ which can be used to determine the spin polarization of the tunneling electrons.[2]

The present experimental results are summa-

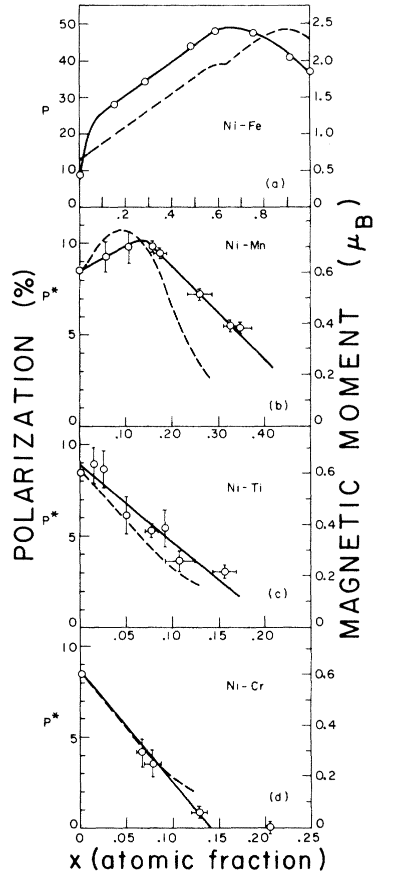

FIG. 1. Measurements of the electron spin polarization P [or P^*, the polarization relative to that of pure Ni (see Ref. 6)] of tunneling currents from Ni alloys shown by points and solid lines versus the atomic fraction of the nonnickel component. The dashed curves show the known values of the saturation magnetic moment n_B of these alloys (see Ref. 7) in Bohr magnetons per atom (right-hand scale).

rized in Fig. 1, which shows the measured electron spin polarization for tunnel junctions in which the ferromagnetic films were nickel alloys containing different concentrations of Fe, Cr, Mn, or Ti. The points and solid lines are the measured polarizations of the alloy junctions. The dashed curves are the accepted values[7] of the saturation magnetic moment of similar alloys at $T \approx 0$. In Fig. 1(a) the variation of P as a function of Fe concentration is rather similar to that of the known behavior of the magnetic moment. In particular in the compositional range of 10 to 60 at.% Fe, both P and n_B increase linearly with the Fe concentration. In this range Fe and Ni form a continuous series of solid solutions and the magnetic anisotropy is rather small. It may be that structural defects, which are always present in the thin films, are less important in this range.

Figure 1(b) shows the values of P^* (the polarization relative to that of pure Ni, see Ref. 6) and n_B for Ni-Mn alloys.[7] The initial increase of n_B and its later decrease are reflected closely in the values of P^*. The fact that n_B drops more rapidly than P^*, particularly at large concentrations, perhaps reflects the higher inhomogeneity of the thin films. Figures 1(c) and 1(d) show the equivalent measurements of P^* for Ti and Cr, respectively. Both have a monotonic decrease which is only slightly less rapid than the decrease for n_B.[7] For each of these alloys the data summarized by Fig. 1 show the close correlation between the magnetic moment and the electron spin polarization. The alloy films were formed by standard evaporation methods. In preliminary experiments with Mn and Ti alloys,[5] the films were prepared by flash evaporation of the intimately mixed powders which were slowly fed into a hot tungsten boat. This technique led to considerably reduced values of P even for pure Ni, an effect which presumably was assoicated with the poor vacuum conditions during extremely rapid evaporation. In the present experiment the alloy films of Ni and Mn, Cr, or Ti were made by co-evaporation of the two elements using two separate quartz-crystal monitoring systems to obtain the desired composition. The Fe-Ni alloy films were formed by evaporating a known composition from a tungsten boat at a known temperature. From the vapor pressure of the elements and the known activity coefficients of Fe and Ni the composition of the deposited film could be accurately calculated.[8] The composition of the deposited films was also measured by x-ray fluorescence. Errors in composition were probably less than 3% except where otherwise shown. During a single evaporation four alloy junctions and four nickel junctions were made on each of four glass substrates. From these 32 junctions we selected those with the best tunneling characteristics. Selected junctions had a leakage current at $V=0$ of less than 1%. The conductance versus voltage was measured with a four-terminal circuit arrangement.[9] Three or four junctions of the Ni and of the alloy films were measured from each evaporation (often on the same substrate). The random error as judged by the variations of P measured on junctions from the same evaporation was approximately $0.02P$. A variation of about ± 1.5 in P (as given in percent) was found between different evaporations of the same nominal concentrations. These latter variations are probably attributable to the effect of different evaporation conditions on the ferromagnetic films, although variations of the thickness and spin-orbit scattering of the Al films may contribute. For small amounts of impurity in Ni, the normalization procedure of using P^* tends to eliminate these sources of error. The values of P given here are not corrected for the effect of spin-orbit scattering in the Al films. This correction, whose exact value is now being determined, will multiply all the values of P by a factor of about 0.9, but leave the relative values of P unchanged.

The above experimental results agree approximately with those on polarization of electrons photoemitted from Ni, Co, and Fe films.[4] As yet no photoemission results have been published on alloy films; such measurements would be of considerable interest. Recent photoemission measurements on Ni single crystals[10] gave negative polarization for energies within 0.05 eV of E_F. However, since the work function changed by 0.11 eV during these measurements, it is hard to judge the validity of these threshold measurements, which must be exceedingly sensitive to surface conditions. However, these results do emphasize the need for single-crystal tunneling measurements, which are very well suited to probe the region near E_F. Regarding the polarization of field-emitted electrons,[11] results to date are complicated by the extreme sensitivity to surface contamination and emission sites,[12] and it seems likely that firm results have not yet been obtained.

From the theoretical side Hertz and Aoi[13] have presented arguments to explain the tunneling results and have derived values of P for Ni, Co, and Fe close to the measured values. Anderson[14] and Doniach[15] have attempted to explain the sign of the polarization in photoemission. Fulde, Luther, and Watson[16] emphasized the importance of surface effects. However, as yet there is no generally accepted explanation for all the results. In any case the observed correlation between P and n_B puts a strong constraint on models of ferromagnetism in the 3d metals.

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