Measurement of the transport spin polarization of FeV using point-contact Andreev reflection

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The $Fe_{1-x}V_x$ alloy system exhibits the lowest known Gilbert relaxation rate of any ferromagnetic metal or binary alloy with G = 35 MHz at x = 27% V. Low relaxation rates are of particular interest in modern spin electronic applications involving spin torque. The transport spin polarization of a series of sputtered epitaxial $Fe_{1-x}V_x$ samples was measured using point contact Andreev reflection. Values of the transport spin polarization agree well with those measured for pure Fe and are independent of composition. The results indicate that the substitution of up to 50% of V for Fe does not reduce the spin polarization in the alloy.
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Spin-momentum transfer (SMT) is the coherent transfer of angular momentum carried by a carrier's spin to influence the direction of magnetization of a second layer when a current is driven through a magnetic metal–non-magnetic spacer–magnetic metal tri-layer structure. The spacer is commonly a tunnel barrier material such as Al_2O_3 or MgO but can also be a metal. Current from one ferromagnet—considered the pinned layer—carries spin-angular momentum that acts on the second, free layer to cause rotation of the magnetization. Parallel and anti-parallel moment orientations are induced with currents in the forward or reverse direction (the sign is material-dependent). The free layer magnetization is aligned in a particular spatial direction by intrinsic and geometric anisotropies. SMT is being explored for use in magnetic random access memory (MRAM) and tunable oscillators.

For MRAM, the critical current necessary to cause switching of the free-layer orientation is

$$I_c = \frac{2e}{\hbar}\frac{\alpha MV}{g(P,\eta)}B_{eff}, \tag{1}$$

where α is the rate of spin damping—classically measured using ferromagnetic resonance methods, *M* is the magnetization, *V* the volume of the free layer, $g(P,\eta)$ is the spin torque efficiency deriving from the spin polarization of the magnetic layers and relative angle (η) between magnetizations, and B_{eff} is the summation of intrinsic and shape anisotropy fields and the externally applied field.[1] Ferromagnetic resonance measurements have shown for $Fe_{1-x}V_x$ the composition at x = 0.27 has a homogenous loss rate of 35 MHz, a value well-below the loss rate of Fe whiskers or epitaxial Fe films (57 MHz) and the previously determined minimum value.[2] The low loss rate makes this material attractive for use in spin-momentum transfer devices. The system $Fe_{1-x}V_x$ forms in the bcc phase over the full compositional range, making it compatible with MgO as a tunnel barrier, and it is expected to benefit from band-matching and spin-filtering inherent in the Fe/MgO/Fe (001) epitaxial architecture.

Even with spin-filtering, it is beneficial that *P* is as large as possible to obtain low J_c for such devices. Soulen *et al.*[3] have shown that *P* can be determined from the current-voltage (I-V) characteristics of junctions formed by a superconducting tip with a ferromagnetic material. The I-V response due to Andreev reflection at the interface[4] is quite different for completely spin-polarized and non-polarized currents.[5] Point contact Andreev reflection (PCAR) has been used to measure the transport spin polarization of a large variety of magnetic systems.[3,6–16] Experimental values of *P* are extracted from the data using a modified Blonder, Tinham, Klapwijk (BTK)[17] model of Andreev reflection at a normal metal/superconductor interface and generalized to include a spin-polarized normal metal.[18]

In this letter we report values of *P* of epitaxial thin films of $Fe_{1-x}V_x$ determined from PCAR measurements and compare them with the results of band structure calculations. Our experimental results show that P is insensitive to composition x in $Fe_{1-x}V_x$ at 1.6 K. While we cannot evaluate the behavior at room temperature directly, we expect that transport spin polarization would also be insensitive to x at room temperature. The existing evidence from spin-polarized tunneling in magnetic tunnel junctions shows that P is nearly temperature-independent in epitaxial Fe near the Fe/MgO interface, with P(300 K)/P(4 K) = 0.93.[19] We expect the same to be true of epitaxial MgO/$Fe_{1-x}V_x$ for temperatures significantly below T_c. The measured room-temperature moments for this film series, from VSM, are consistent with the expected (low-T) Slater-Pauling variation and show no decreasing room-temperature moment due to decreasing T_c with increasing concentration x.[2]

Epitaxial MgO(001)/50 nm $Fe_{1-x}V_x$(001) samples were deposited by UHV co-sputtering according to conditions detailed in previous reports.[2,20] Base pressures for the depositions were 2×10^{-9} Torr, Ar pressures were 3 mTorr, and substrate temperatures were 200 °C. A linear reduction in room-temperature magnetic moment $4\pi M$ and monotonic reduction in magnetocrystalline anisotropy constant

K_2, to null at x = 44%, with increasing V concentration, x, have been observed in the samples studied, as shown in Ref. 2. Characterization of element-specific moments on Fe and V sites by X-ray magnetic circular dichroism (XMCD), for similarly prepared polycrystalline films, has been shown by Guan *et al*.[21] Additional samples were grown at NRL for x = 0.10, 0.20, and 0.30 using the same pressure and temperature conditions. We additionally performed an Ar sputter-cleaning of the x = 0.23 sample to determine whether surface oxidation affects the measured polarization. No difference was found in P as a result of this cleaning procedure.

The superconducting tip (Sn or Nb) used in this study was driven by a micrometer mechanism capable of moving the point linearly by 100 μm per revolution. While it is not possible to determine the microscopic nature of the contact in this apparatus, our previous work has shown that the values of P obtained agree with those obtained from other techniques.[3,6] Measurements of the current-voltage (I-V) and differential conductance $G = (dI/dV)$ characteristics were made using a conventional four-terminal probe arrangement. Both the point contacts and samples were immersed in a liquid helium bath between 4.2 K and 1.5 K with conductance data obtained by a standard ac lock-in technique at a frequency of 2 kHz. Further details of the measurement technique can be found in Refs. 3 and 6. We studied seven members of the $Fe_{1-x}V_x$ series ($0 \le x \le 0.52$).

Ten or more conductance curves, corresponding to different contacts, have been measured for each sample. The data were analyzed by fitting each individual data set with the modified BTK model that included the correct renormalization for the half-metallic case.[18] The fitting routine includes corrections for non-linear conductance above the superconducting gap as well as "spreading" film resistance (which were negligible due to the low resistance of the samples). With this procedure the only two parameters that are varied in the fitting program, which uses a least square fit routine, are the spin polarization P and the strength of the BTK delta-function, Z. In general one would expect P to be independent of Z, unless there is strong spin-flip scattering at the interface. One example is CrO_2 with the antiferromagnetic Cr_2O_3 surface layer.[9] If such a layer is present, the measured P values will likely depend on Z.

The temperature is taken to be the measured physical temperature of the film, while the value of the superconducting gap was taken from the BCS temperature dependence and known to be very close to the experimental values for Sn and Nb. In order to ensure that the extracted values of P were robust, we performed the analysis for several samples using elevated effective temperatures (up to 4 K) along with the corresponding energy gaps and found no changes to within the reported uncertainties.

Figure 1 shows an example of normalized conductance data for one of the samples obtained with a Sn tip, while Figure 2 shows an example of the PCAR data for a sample obtained with a Nb tip. Both measurements were obtained at a temperature of ~1.6 K. The solid line is the modified BTK fit to the data. Table I gives a summary of the results for all of the samples. The values of P vary from 0.4 to 0.53. There was no dependence of P on Z in our data.

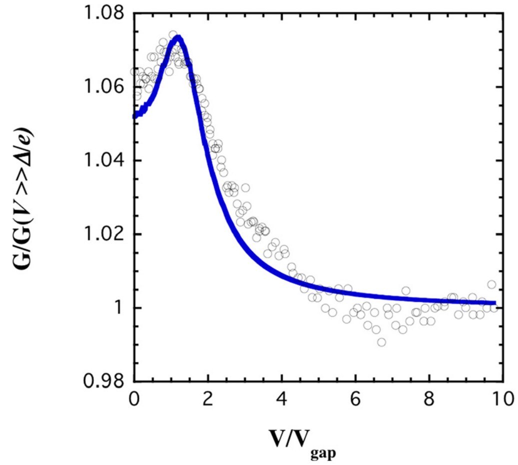

FIG. 1. Example of a conductance curve (normalized for $V \gg \Delta/e$) with a Sn tip taken at 1.7 K. The solid curve is the modified BTK theory fitted values: $P = 47\%$ and interface barrier strength $Z = 0.005$.

The transport spin polarization has to be distinguished from the most common definition of the spin polarization, P_0, of the density of states (DOS), $P_0 = \{N_\uparrow(E_F) - N_\downarrow(E_F)\}/\{N_\uparrow(E_F) + N_\downarrow(E_F)\}$, where $N_{\uparrow\downarrow}(E_F)$ is the density of states for the majority (minority) bands at the Fermi level. In general, spin-polarization in the PCAR experiments can be represented by the expression

$$P_n = \frac{I_\uparrow - I_\downarrow}{I_\uparrow + I_\downarrow} = \frac{\langle N(E_F)v_F^n\rangle_\uparrow - \langle N(E_F)v_F^n\rangle_\downarrow}{\langle N(E_F)v_F^n\rangle_\uparrow + \langle N(E_F)v_F^n\rangle_\downarrow}, \qquad (2)$$

where v_F is the Fermi velocity of the majority (minority) spins and n is either 1 or 2, depending upon whether conduction in the contact is in the ballistic or diffusive regime, respectively.[18] In the special case of a spatial tunnel barrier[22] the spin polarization measured by the tunneling technique is reduced to Eq. (2) with $n = 2$. It is important to keep in mind that the transport spin polarization is relevant for spintronics applications and may be dramatically different from P_0.[9]

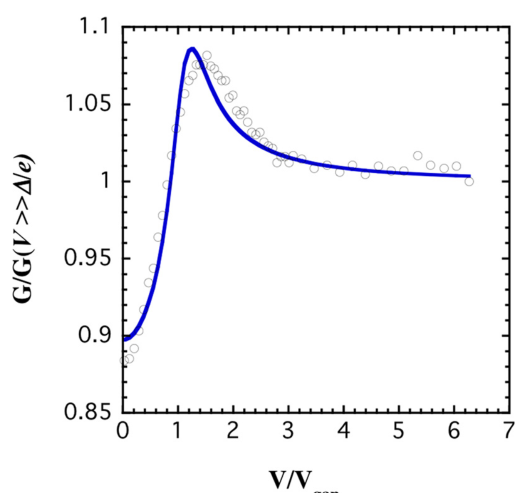

FIG. 2. Example of a conductance curve for sample 264 with a Nb tip taken at 1.6 K, normalized for $V \gg \Delta/e$. The solid curve is the modified BTK theory fitted values: P = 51% and barrier strength Z = 0.17.

TABLE I. Summary of the spin-polarization results for the FeV samples studied. *P* is the average spin-polarization.

Sample	V (%)	Tip	P	Contacts
262 (as received)	0	Nb	0.49 ± 0.04	19
263 (as received)	10	Nb	0.50 ± 0.05	10
264 (sputter cleaned)	23	Sn	0.41 ± 0.06	3
264 (sputter cleaned)	23	Nb	0.43 ± 0.06	10
264#2 (sputter cleaned)	23	Nb	0.49 ± 0.02	6
266 (as received)	42	Nb	0.40 ± 0.06	14
267 (as received)	52	Nb	0.52 ± 0.03	15
kb091230A (as received)	30	Nb	0.51 ± 0.03	17
kb091231B (as received)	40	Nb	0.53 ± 0.05	14

In order to address theoretically the effect of vanadium-iron mixtures upon the spin polarization, as measured in the Andreev reflection experiment, we have performed first principles density functional theory calculations, using the linearized augmented plane wave (LAPW) code WIEN2K within the generalized gradient approximation of Perdew, Burke, and Ernzerhof. These calculations consisted of self-consistent $2 \times 2 \times 2$ supercell calculations, where the size of the supercell is in units of a single body-centered cubic cell. Since the lattice constant of bcc vanadium is somewhat larger than that of bcc iron, we performed these calculations at an equivalent, i.e., single bcc cell, lattice constant of 2.90 A, intermediate between the Fe lattice constant of 2.87 A and that of vanadium (3.02 A), and for simplicity used the same lattice constant for all calculations. Calculations were run for sixteen separate sixteen-atom supercells with the vanadium/iron content varied in increments of one atom. A very fine k-mesh of 816 points in the irreducible Brillouin zone was used and the LAPW sphere radius was 2.36 Bohr radii for all atoms. Calculations were run to self-consistency to an energy convergence threshold of better than 1 meV per 16 atom unit cell. From these converged calculations, the three different measures of spin polarization, as formulated by Mazin,[18] were extracted.

As expected, the model predicts that the saturation moment decreases monotonically to zero with V concentration (at $x \sim 70\%$) consistent with the theoretical work of Johnson *et al.*[23] and experimental work in Ref. 2. Magnetization data obtained from the films are in very good agreement with the calculations (Figure 3). The calculated values of the three polarizations are shown in Figure 4. As expected, there are significant differences in the behaviors of the polarizations. Perhaps unexpectedly, the results show negative polarizations below $x \sim 0.25$ and rather robust until the saturation moment disappears at $x \sim 0.70$. The negative polarization is consistent with the observation of inverse current perpendicular to plane giant magnetoresistance (CPP-GMR) in $Fe_{1-x}V_x$/Au/Co spin-valves.[24–26]

Since the PCAR measurement is not sensitive to the sign of the polarization, the results must be compared to the absolute value of the calculated P (Figure 5). It is apparent that the polarization is fairly constant over the full compositional series $0 < x < 0.5$, near the $P \sim 47\%$ reported previously for pure Fe.[3] Remarkably, there is no evidence of the predicted inversion in spin polarization P as a function of V

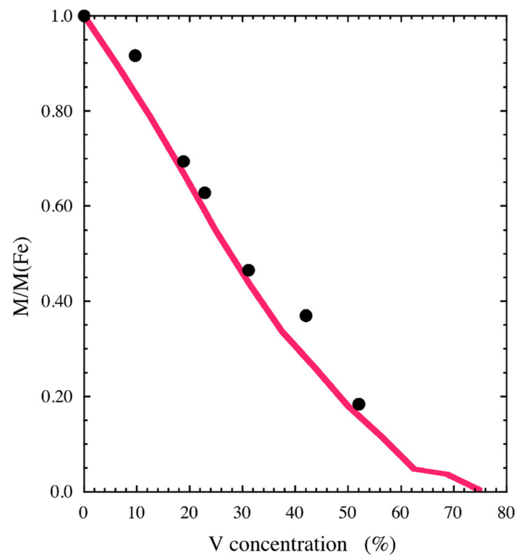

FIG. 3. Normalized saturation moment as a function of V concentration. The solid line is the behavior predicted by the band theory calculation.

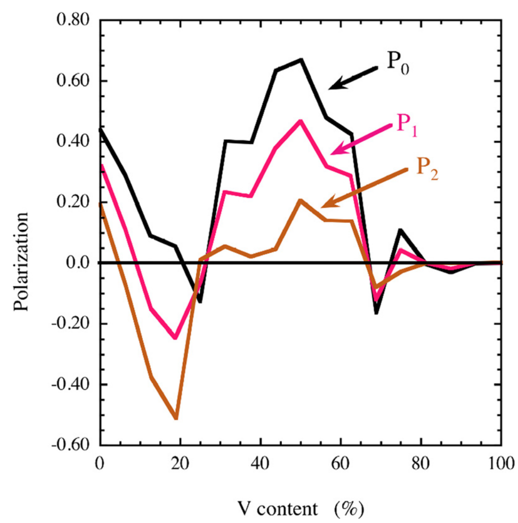

FIG. 4. Calculated predictions for the density of states polarization (P_0), the ballistic polarization (P_1), and the diffusive polarization (P_2).

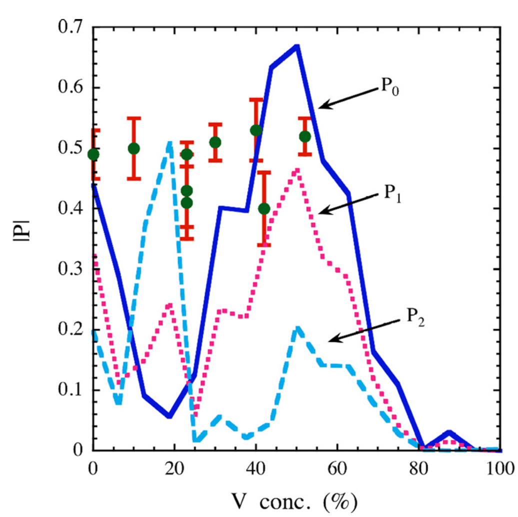

FIG. 5. Polarization data plotted with the calculated predictions for absolute values of the density of states polarization (P_0), the ballistic polarization (P_1), and the diffusive polarization (P_2).

composition x in our PCAR measurements. The dramatic variation of |P| as a function of x, predicted in each of the diffusive, ballistic, or DOS polarization calculations, clearly does not reproduce the experimental data in Figure 5. We understand the suppression of this behavior to be the result of site disorder, necessarily present in the random solid-solution epitaxial films and not considered in the calculations, which approximate the alloys by using ordered supercells, with attendant periodicities not present in the actual material. Spin polarization is known to be sensitive to site disorder in some materials, such as the NiMnSb Heusler alloy.[27]

In conclusion, we have done point contact Andreev reflection measurements of the spin polarization of the itinerant ferromagnet $Fe_{1-x}V_x$. The transport spin polarization on average is 40%–50%. Our experimental results are consistent with the maximum calculated P for the ballistic case using first principles density functional theory but do not agree with the predicted P(x) dependence. This disagreement is likely due to lattice disorder.

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