Magnetic field-enhanced spin filtering in NbN-DyN-NbN spin-filter tunnel junctions

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Spin filter tunnel junctions are based on selective tunneling of up and down spin electrons controlled through exchange splitting of the band structure of a ferromagnetic insulator. Therefore, spin filter efficiency can be tuned by adjusting exchange strength of the tunnel barrier. We have observed that magnetic field and bias voltage (current) can be used to regulate exchange strength and consequently spin-filter efficiency in tunnel junctions with ferromagnetic DyN tunnel barrier. We have found that DyN forms high quality tunnel barrier of height Φ0 = 60 meV with NbN electrodes. We obtained ~37% spin polarization of tunneling electrons at 11 K due to a small exchange splitting ΔE∥ = 5.6 meV of the barrier height Φ0. We observed enhancement in Curie temperature TCurie of DyN with magnetic field which hint field-induced enhanced magnetism in DyN. In presence of an applied magnetic field barrier height can further split due to magnetic field dependent exchange energy ΔE∥(H). The spin filter efficiency in these tunnel junctions can be increased up to ~87% with magnetic field. Electric and magnetic field tuned spin-filter efficiency of DyN tunnel barrier gives opportunity for practical application of these devices with additional functionality.

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The creation, injection and transport of spin-polarized current are the three basic steps in all spintronics devices. Instead of relying on conventional ferromagnets for creation, fully spin polarized currents can also be generated using spin-filter tunneling of electrons through a ferromagnetic tunnel barrier. Starting from unpol arized electrons of a non-magnetic electrode, spin-up and spin-down electrons passing through a spin-filter tunnel barrier can be filtered due to the difference in tunnel barrier heights for the two spin channels. The difference in barrier height appears due to exchange splitting of the band structure, which leads to the conduction band minima and valance band maxima at different energies for majority and minority spin electrons. Spin filter tunnel barrier can also solve impedance mismatch problem with semiconducting counter electrodes facilitating in spin injection from even a non-magnetic metal into a semiconductor. Many ferromagnetic insulators have been tested for their spin filtering property: the Eu chalcogenides including EuS, EuSe, EuO, etc. have shown high spin filtering property; the Eu chalcogenides including EuS, EuSe, EuO, etc. have also received a lot of attention due to their higher Curie temperature TCurie. As 4f moments in Re atoms are highly localized, the exchange interaction in these compounds is mediated by induced moments in Re − d and N − p orbitals. Most widely studied ferromagnetic ReN is GdN which shows the highest Curie temperature TCurie ∼70 K. In this paper, we have focused on one of the poorly explored rare earth nitrides DyN: a ferromagnetic semiconductor with TCurie ∼35 K. DyN has two more 4f electrons than the half-filled GdN. It has the highest saturation magnetization ~10µB/Dy among the series. Theoretical calculations show a small indirect Γ-X gap ∼0.34 eV and a minimum direct gap of ~1.17 eV at X[20]. Experimentally DyN has been shown to be a semiconductor with an optical gap of ~1.2 eV[21]. Besides ferromagnetism, magnetocaloric effect has also been observed in DyN[22]. This makes the spin filtering property of DyN tunnel barrier at higher magnetic field very interesting. Recently, we have made an extensive study of the tunneling property of NbN-DyN-NbN tunnel junctions with different thickness of DyN[23]. We found a crossover from diffusive to tunneling transport as the DyN thickness is made smaller than ~4 nm.

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In this paper, we show that the tunneling properties of DyN junction are strongly affected by magnetic field. We demonstrate that splitting of the tunnel barrier height for spin-up and down electron can be further increased with magnetic field. We have explained our data with simple tunneling model of Simmons considering different conductance channel for up-spin and down-spin electrons.

NbN-DyN-NbN trilayer films were deposited on 5 × 5 mm² Si/SiO₂(250 nm)/MgO(10 nm) substrates by reactive dc sputtering. The deposition was done with a pressure of ~1.5 Pa at room temperature in a UHV chamber equipped with multiple layer deposition. Superconducting NbN was deposited with 28% Ar-N₂ gas mixture at 100 W sputtering power. Whereas 8% Ar-N₂ gas mixture with lower ~16 W sputtering power was used for DyN. The details of deposition condition is described in reference[23]. Trilayers with 50 nm NbN and 3 nm DyN barriers were used for tunnel junction fabrication. Eight tunnel junctions of dimension 7 × 7 µm² were fabricated on each substrate by optical lithography in conjunction with Ar-ion milling and CF₄ plasma etching. Electrical characterization of the tunnel junctions were done in a four-probe configuration in a closed-cycle He refrigerator from Cryogenic Limited. Although all the junctions behaved in a similar way, for consistency we have shown measurements done on one junction only in this paper. Magnetization measurements were performed in a Quantum Design SQUID magnetometer.

The tunneling nature of the junctions was confirmed through $I-V$ measurements at different temperatures. Assuming that at 50 K the exchange splitting of the barrier $E_{ex} = 0$, the Simmons model was fitted to the $I-V$ curve and barrier height $\Phi_0 = 60$ meV and width $d = 3.3$ nm were found (see Supplementary Information[24]). Below $T_{Curie}$, the conduction band is split by the ferromagnetic exchange interaction leading to a lower barrier height $\Phi_\uparrow = \Phi_0 - E_{ex}$ for up-spin and higher $\Phi_\downarrow = \Phi_0 + E_{ex}$ for down-spin electrons. The spin filtering efficiency is usually given by $P = \frac{J_\uparrow - J_\downarrow}{J_\uparrow + J_\downarrow}$; where $J_\uparrow$ and $J_\downarrow$ are spin-up and spin-down current, respectively[2]. As per Wentzel-Kramers-Brillouin (WKB) approximation the tunneling current density for each spin sign exponentially depends on the relevant barrier height $\Phi_\uparrow$ (or $\Phi_\downarrow$) and can be written as $J_{T(\downarrow)} \propto \exp \left(-\frac{\Phi_\downarrow}{d} \sqrt{\frac{2m}{\hbar^2}}\right)$; where $d$ is the thickness of the tunnel barrier and $m$ is the electron mass. In the presence of a magnetic field the barrier height can split further by Zeeman energy, $E_Z = g \mu H$. Where $g$ is the Landé g-factor and $\mu$ is the magnetic moment per formula unit of DyN. We have used $g = 2$ and $\mu = 10 \mu_B$ for our calculations. Besides Zeeman splitting magnetic field dependence of

![FIG. 1: (Color online) Temperature dependence of resistance of a NbN-DyN(3 nm)-NbN tunnel junction with zero (red) and 1 T (blue) magnetic field measured using a current $I = 1 \mu A$.](Image)

![FIG. 2: (Color online) (a) Schematic illustration of the spin filter mechanism in presence of magnetic field. Spin-up and spin-down bands are further split by magnetic field dependent exchange energy $E_{ex}(H)$ (shown in orange color). (b) Expected spin-filter efficiency $P$ for different thickness $d$ of the tunnel barrier with magnetic field $H$ at low bias voltage($V=0$). The spin-filter efficiency was calculated for a spin filter barrier with barrier height $\Phi_0 = 60$ meV and exchange field $E_{ex} = 5.6$ meV (see text for details).](Image)

vides proof that spin filtering is present in our junctions; spin filtering efficiency can be determined from $R(T)$ using the method described in the reference[10], and gives a spin filtering efficiency $P \sim 37\%$ at 11 K in zero magnetic field. This is quite small compared to $P \sim 90\%$ at the critical temperature of NbN in NbN-GdN-NbN spin filter tunnel junctions. When an in-plane magnetic field is applied we found that the spin filter efficiency increased from $P \sim 67\%$ at 1 T to $P \sim 87\%$ at 5 T. The change in slope in the $R(T)$ ($dR(T)/dT > 0$ to $dR(T)/dT < 0$) can be used as an indicator of $T_{Curie}$. Fig. 1 shows that this is significantly enhanced at 1 T.
exchange energy $E_{\text{ex}}(H)$ also has to be considered. In the Curie regime (i.e., $g\mu_B B < k_B T$) magnetization $M(H) \propto H^{\frac{3}{2}}$. Considering $E_{\text{ex}}(H) \propto M(H)$, we can write $E_{\text{ex}}(H) = E_{\text{ex}}(0) + \alpha_s H$. Here $\alpha_s$ is a temperature dependent constant related to strength of exchange interaction between moments on Dy atoms. Fig. 2(a) shows a schematic illustration of the spin-filter mechanism in the presence of magnetic field. Additional spin splitting of the tunnel barrier due to magnetic field dependent $E_{\text{ex}}(H)$ is shown in orange color. Consequently, the spin filter efficiency at very low applied bias voltage ($V \rightarrow 0$) in presence of magnetic field can be written as:

$$P = \frac{e^{-\frac{\hbar}{2}\sqrt{2m[\Phi_0 - E_{\text{ex}}(H) - E_z]}} - e^{-\frac{\hbar}{2}\sqrt{2m[\Phi_0 + E_{\text{ex}}(H) + E_z]}}}{e^{-\frac{\hbar}{2}\sqrt{2m[\Phi_0 - E_{\text{ex}}(H) - E_z]}} + e^{-\frac{\hbar}{2}\sqrt{2m[\Phi_0 + E_{\text{ex}}(H) + E_z]}}}$$

(1)

The exchange energy $E_{\text{ex}}$ can be calculated with a known value of the spin-filter efficiency using Eq.[1]. At 11 K using $P \sim 37\%$ for our tunnel junction we found $E_{\text{ex}} = 5.6$ meV for $H = 0$. The spin-filter efficiency is usually determined by the ratio $E_{\text{ex}}/\Phi_0$ which is only $\sim 0.09$ in our tunnel junction. For comparison, in EuO spin filter junctions $E_{\text{ex}}/\Phi_0 \sim 0.3$ which leads to a very high spin-filter efficiency $\sim 98\%$[2]. Fig. 2(b) shows the calculated spin-filter efficiency of a tunnel barrier with $\Phi_0 = 60$ meV and $E_{\text{ex}} = 5.6$ meV at different magnetic fields with $\alpha_s = 0.004$. Clearly very high spin-filter efficiency ($\sim 100\%$) can be obtained with smaller magnetic field in thicker tunnel barrier. Therefore, magnetic field can be used to tune spin filter efficiency in our tunnel junctions.

![Fig. 3](image3.png)

Fig. 3 shows the magnetoresistance ($MR = (R(H) - R(0))/R(0)$) of a NbN-DyN(3 nm)-NbN tunnel junction at different temperatures. At 100 K negligibly small $MR$ was found. As temperature is decreased $MR$ is found to increase rapidly with magnetic field. At 15 K $MR \sim 45\%$ was found for 5 T magnetic field. The MR showed strongly nonlinear dependence with magnetic field which becomes more pronounced as temperature is decreased.

As Curie temperature $T_{\text{Curie}} \sim 35$ K, the magnetization of the DyN barrier increases along with increase in spin-filter efficiency as the junction is cooled below $T_{\text{Curie}}$. Therefore, the nonlinear nature of MR can be understood by considering magnetic field dependence of barrier heights $\Phi_{t(\downarrow)}$. Field dependence of magnetization of the NbN-DyN(3 nm)-NbN trilayer film measured at 13 K is shown in the inset of Fig. 3. The film was deposited at the same time as the trilayer from which device is made. We also measured MR with different value of bias current. Fig. 4 shows the field dependence of $MR(H)$ of the tunnel junction measured at 11 K (just above the $T_C$ of NbN) with different bias current. For low bias current $\sim 10 \mu A$ very large $MR \sim 45\%$ was found with a magnetic field of 5 T. The $MR$ showed a prominent nonlinear behavior with magnetic field $H$. The Magnetoresistance was found to reduce significantly when bias current was increased to $\sim 1$ mA.

![Fig. 4](image4.png)

We now try to explain the magnetic field and bias current (voltage) dependence of resistance in our junctions with tunneling model. Similar to Mott’s two current model[26], we assume the spin currents for spin-up and spin-down electrons are independent of each other. Therefore, the total resistance of our tunnel junction can be written as, $G = G_\uparrow + G_\downarrow$; where $G_\uparrow$ and $G_\downarrow$ are conductances for up-spin and down-spin electrons, respectively. In the low voltage limit ($V \rightarrow 0$) the conductances $G_\uparrow$ and $G_\downarrow$ can be calculated from Simmons’ model[27] as:
FIG. 5: Calculated $MR$ as a function of magnetic field for different magnetic strength $\alpha_s$ of the tunnel barrier (see text for details).

$$G_{\uparrow(\downarrow)} = \frac{3\sqrt{2m\Phi_{\uparrow(\downarrow)}}}{2d} \left(\frac{e}{\hbar}\right)^2 \exp\left(-\frac{2d}{\hbar}\sqrt{2m\Phi_{\uparrow(\downarrow)}}\right) \tag{2}$$

Where $m$ is mass of the electron and $\hbar$ is Planck’s constant. Here barrier height for spin-up and spin-down electrons are: $\Phi_{\uparrow} = \Phi_0 - E_{ex} - \alpha_s H - E_Z$ and $\Phi_{\downarrow} = \Phi_0 + E_{ex} + \alpha_s H + E_Z$, respectively. Calculated $MR$ for our tunnel junction with $\Phi_0 = 60$ meV and $E_{ex} = 5.6$ meV using Eq[2] is shown in Fig.

5. Clearly one can see that as $\alpha_s$ increases $MR$ also increases rapidly and becomes nonliner with magnetic field. The constant $\alpha_s$ is related to the magnetic strength of the tunnel barrier. Therefore, increases in nonlinearity with temperature as shown in Fig. 3 is related to the increase in magnetic strength of the tunnel barrier.

In conclusion, we have fabricated NbN-DyN-NbN spin-filter tunnel junctions and made an extensive study on the effect of applied bias voltage and magnetic field on the spin-filtering. Using the Simmons model we have calculated barrier height $\Phi_0 = 60$ meV in our tunnel junctions. A small exchange splitting $E_{ex} = 5.6$ meV of the barrier height at 11 K resulted in $\sim 37\%$ spin-filter efficiency in our tunnel junctions. The spin filter efficiency is found to increase to $\sim 87\%$ when magnetic field $H = 5$ T was applied. The increase in spin-filter efficiency can be understood by considering further lowering of the up-spin barrier height by magnetic field-induced increase in exchange energy $E_{ex}(H)$. Our findings are potentially useful for the control of spin polarized super-current in spintronics devices.

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