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Electronic Supplementary Information

Tunneling Magnetoresistance and Light Modulation in $Fe_4N(La_{2/3}Sr_{1/3}MnO_3)/C_{60}/Fe_4N$ Single Molecule Magnetic Tunnel Junctions

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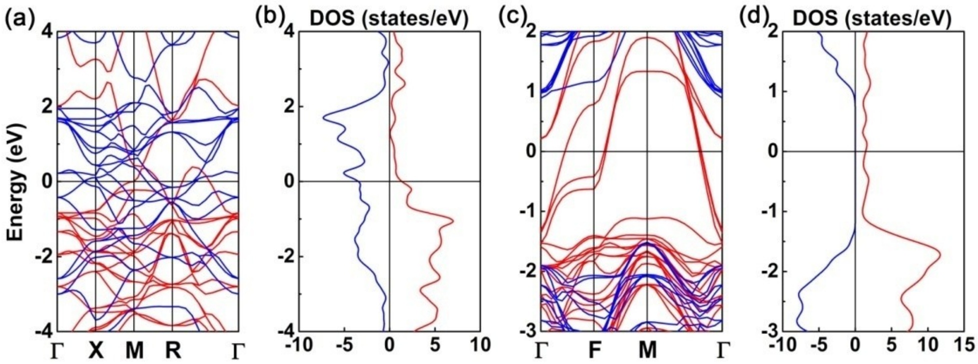

Fig. S1 (a) Band structure and (b) density of states (DOS) of bulk Fe_4N. (c) Band structure and (d) DOS of bulk $La_{2/3}Sr_{1/3}MnO_3$. The red and blue lines distinguish spin-up and spin-down, respectively.

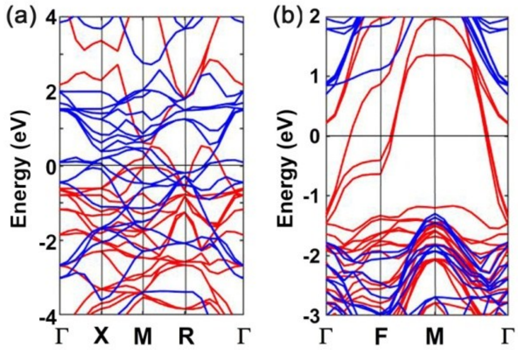

Fig. S2 Band structures of bulk Fe_4N (a) and $La_{2/3}Sr_{1/3}MnO_3$ (b) calculated by Nanodcal package. The red and blue lines distinguish spin-up and spin-down band structure, respectively.

The spin-dependent current was further obtained using the Landauer-Büttiker formula[1]

$$I_\sigma = \frac{e}{h}\int_{\mu_\mathrm{L}}^{\mu_\mathrm{R}} T_\sigma\left(E,V\right)\left[f\left(E-\mu_\mathrm{R}\right)-f\left(E-\mu_\mathrm{L}\right)\right]\mathrm{d}E\,, \tag{1}$$

where e is the electron charge, h is Planck's constant, and $T_\sigma\left(E,V\right)$ is the spin-dependent ($\sigma = \uparrow,\downarrow$) transmission coefficient at a bias voltage $V = V_\mathrm{R} - V_\mathrm{L}$. $f\left(E-\mu_{\mathrm{L(R)}}\right)$ is Fermi-Dirac distribution function related to the chemical potential $\mu_{\mathrm{L(R)}}$ of the left(right) electrode.

The spin injection efficiency (SIE) and tunneling magnetoresistance (TMR) at a voltage are defined as

$$\mathrm{SIE} = \frac{I_\uparrow\left(V\right)-I_\downarrow\left(V\right)}{I_\uparrow\left(V\right)+I_\downarrow\left(V\right)}\times 100\%\,, \tag{2}$$

$$\mathrm{TMR} = \frac{I_\mathrm{PC}\left(V\right)-I_\mathrm{APC}\left(V\right)}{I_\mathrm{APC}\left(V\right)}\times 100\%\,, \tag{3}$$

where I_PC and I_APC are the total currents in PC and APC, respectively. At the equilibrium state, SIE and TMR are calculated from the electron transmittance coefficient ($T(E_\mathrm{F})$) at Fermi level.

For linearly polarized light, the polarization vector can be defined as $\mathbf{e}=\cos\theta\ \mathbf{e}_1 + \sin\theta\ \mathbf{e}_2$, where θ is the angle between the polarization direction and the vector $\mathbf{e}_1$. The photocurrent moving into the left electrode is defined as follows[2, 3]

$$J_L^{(\mathrm{ph})} = \frac{ie}{h}\int(\cos^2\theta \mathrm{Tr}\{\Gamma_L[G_1^{<(\mathrm{ph})} + f_L(G_1^{>(\mathrm{ph})} - G_1^{<(\mathrm{ph})})]\}$$

$$+\sin^2\theta \mathrm{Tr}\{\Gamma_L[G_2^{<(\mathrm{ph})} + f_L(G_2^{>(\mathrm{ph})} - G_2^{<(\mathrm{ph})})]\}$$

$$+\frac{\sin(2\theta)}{2}\mathrm{Tr}\{\Gamma_L[G_3^{<(\mathrm{ph})} + f_L(G_3^{>(\mathrm{ph})} - G_3^{<(\mathrm{ph})})]\})dE\,, \tag{4}$$

where

$$G_1^{>/<(\mathrm{ph})} = \sum_{\alpha,\beta=x,y,z} C_0 N G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a\,,$$

$$G_2^{>/<(\mathrm{ph})} = \sum_{\alpha,\beta=x,y,z} C_0 N G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a\,,$$

$$G_3^{>/<(\mathrm{ph})} = \sum_{\alpha,\beta=x,y,z} C_0 N(G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a + G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a)\,. \tag{5}$$

The polarization vector of elliptically polarized light is **e**=cosϕ **e**$_1$ + i sinϕ **e**$_2$, where ϕ determines its helicity. ϕ=±45° corresponds to right- and left-handed circularly polarized light, then the photocurrent is described as

$$J_L^{(\mathrm{ph})} = \frac{ie}{2h}\int(\mathrm{Tr}\{\Gamma_L[G_1^{<(\mathrm{ph})} + f_L(G_1^{>(\mathrm{ph})} - G_1^{<(\mathrm{ph})})]\}$$

$$+\mathrm{Tr}\{\Gamma_L[G_2^{<(\mathrm{ph})} + f_L(G_2^{>(\mathrm{ph})} - G_2^{<(\mathrm{ph})})]\}$$

$$\pm\mathrm{Tr}\{\Gamma_L[G_3^{<(\mathrm{ph})} + f_L(G_3^{>(\mathrm{ph})} - G_3^{<(\mathrm{ph})})]\})dE\,, \tag{6}$$

where $G_{1,2}^{>/<(\mathrm{ph})}$ is the same as the linearly polarized case and

$$G_3^{>/<(\mathrm{ph})} = \pm i\sum_{\alpha,\beta=x,y,z} C_0 N(G_0^r e_{1\alpha} p_\alpha^* G_0^{>/<} e_{2\beta} p_\beta G_0^a - G_0^r e_{2\alpha} p_\alpha^* G_0^{>/<} e_{1\beta} p_\beta G_0^a)\,. \tag{7}$$

In the above equations, $C_0 = (e/m_0)^2(\hbar\sqrt{\mu_r\varepsilon_r}/2N\omega\varepsilon c)I_\omega$, where m_0 is the bare electron mass, I_ω is the photon flux defined as the number of photons per unit time per unit area, ω is the frequency and c is the speed of light, μ_r is the relative magnetic susceptibility, ε_r is the relative dielectric constant, ε is the dielectric constant, and N is the number of photons. $G_0^{r/a}$ are the retarded and advanced Green's functions without photons, respectively. Meanwhile, $G_0^{>/<}$ are greater and lesser Green's function without photons, respectively. $p_{x,y,z}$ is the Cartesian component of the electron momentum, and $e_{1/2x,y,z}$ is the Cartesian component of the unit vector $\mathbf{e}_{1/2}$, which characterizes the polarization of the light. The normalized photocurrent is written as

$$R = \frac{J_L^{(\mathrm{ph})}}{eI_\omega} . \tag{8}$$

Note that the unit of R is a_0^2/photon where a_0 is the Bohr radius.

References

1. Y. Meir and N. S. Wingreen, *Phys. Rev. Lett.*, 1992, **68**, 2512–2515.
2. Y. Xie, L. Zhang, Y. Zhu, L. Liu and H. Guo, *Nanotechnology*, 2015, **26**, 455202.
3. Y. Xie, M. Chen, Z. Wu, Y. Hu, Y. Wang, J. Wang and H. Guo, *Phys. Rev. Appl.*, 2018, **10**, 034005.