Colossal spin fluctuation in a molecular quantum dot magnet

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in collaboration with

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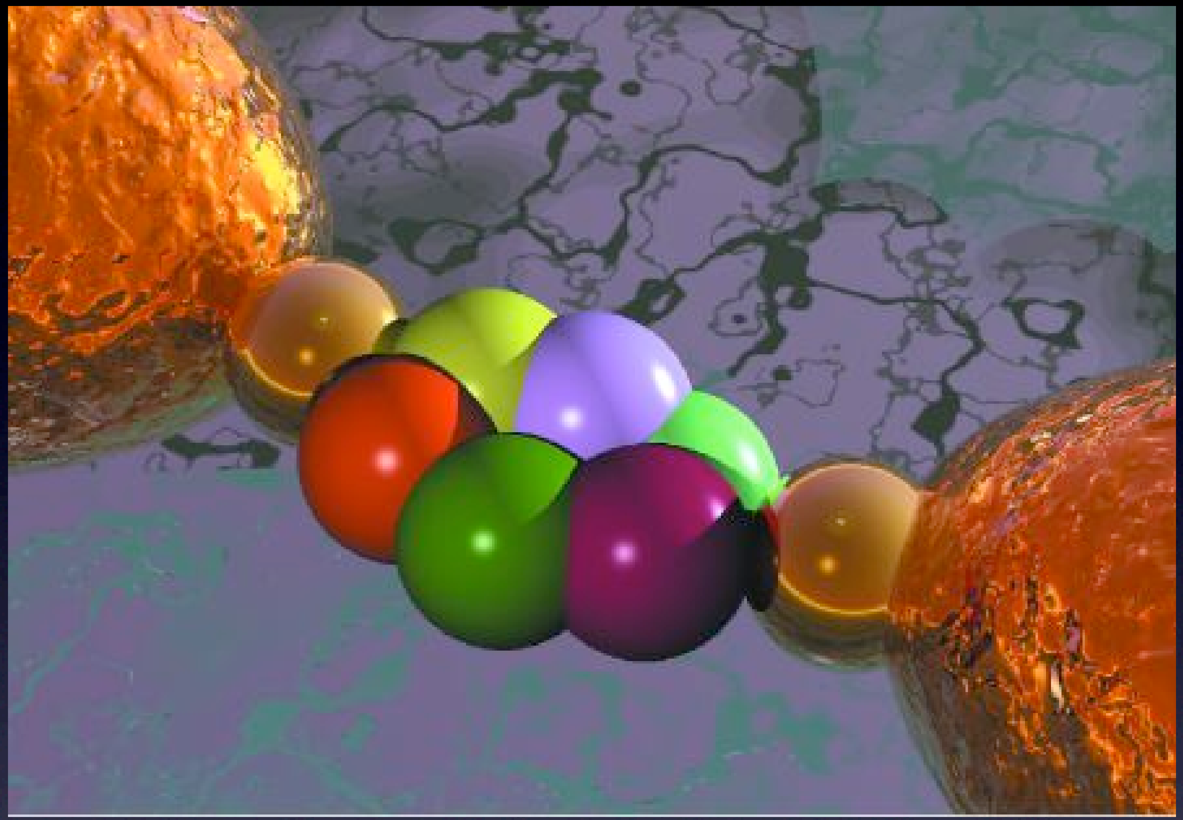
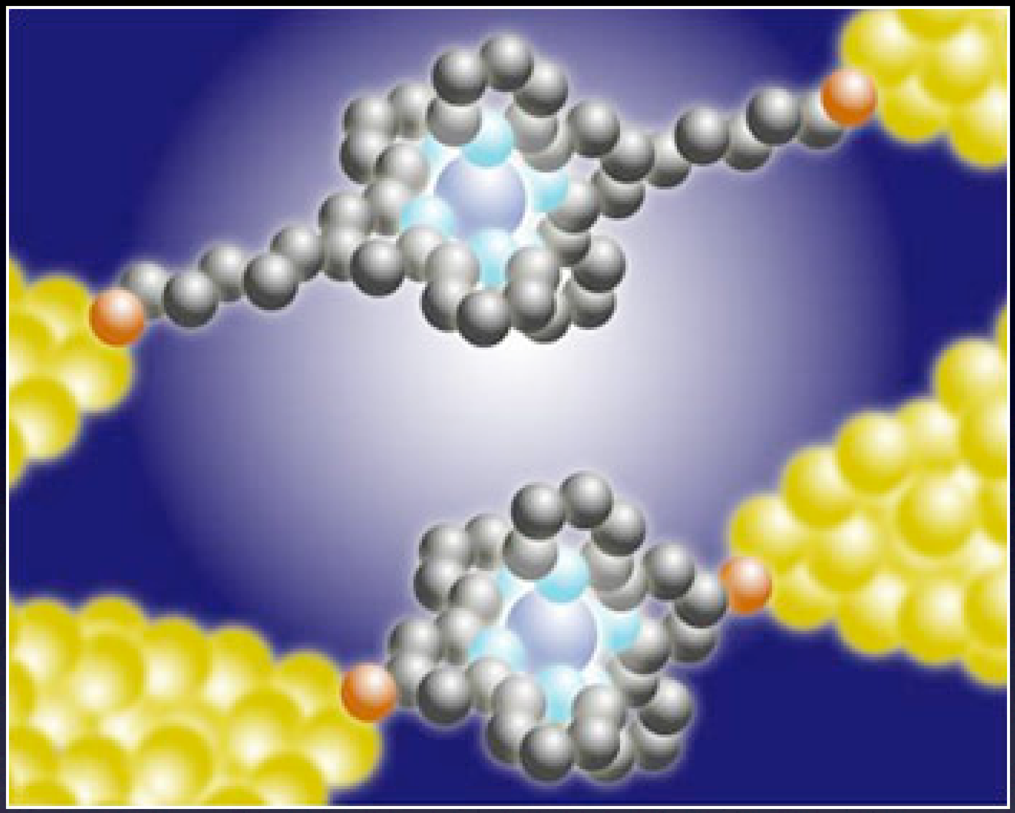
The system

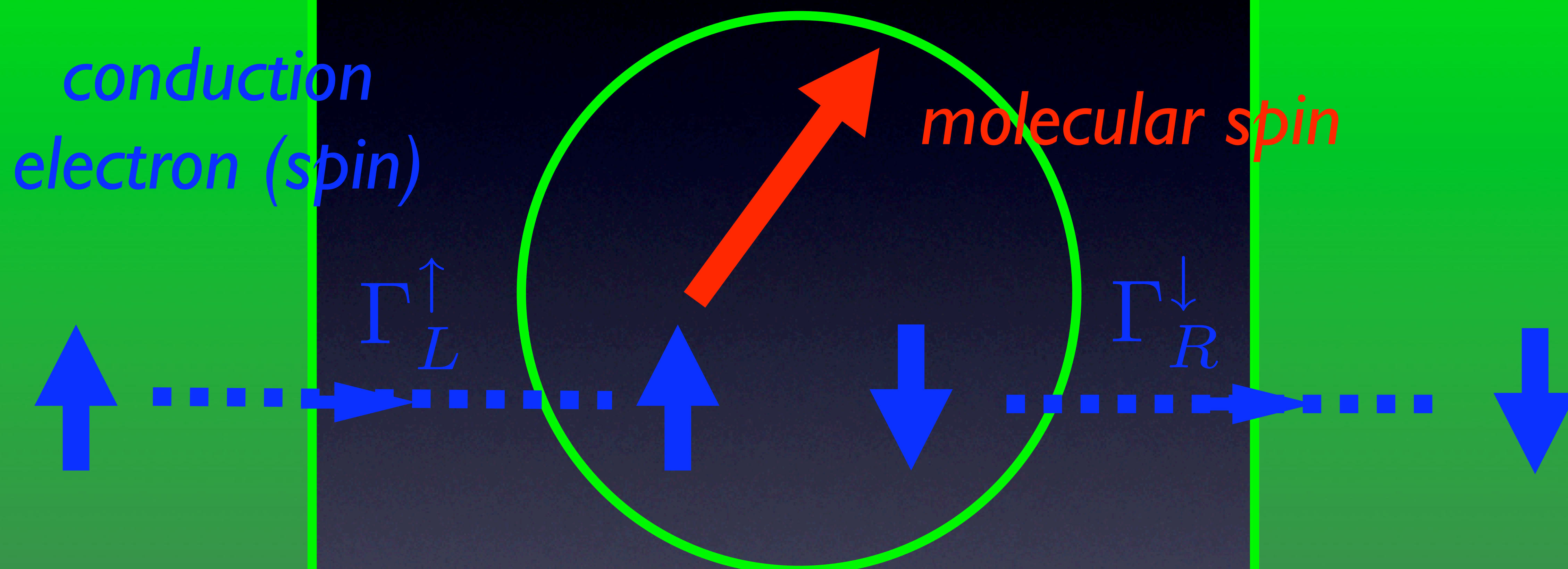
A molecular quantum dot magnet





- Molecules play the role of a quantum dot
- Molecules have their own spin, coupled to conduction electrons' spin via an exchange interaction
- Our molecular quantum dot magnet is coupled to ferromagnetic leads

Model for a Molecular quantum dot magnet



$$H_{\rm dot} = \epsilon_{\rm dot} \sum_{\sigma} n_{\sigma} + U n_{\uparrow} n_{\downarrow} + H_{\rm spin}$$

$$H_{\rm spin} = \begin{cases} 0 & (n = 0) \\ g\vec{\sigma} \cdot \vec{s} & (n = 1) \end{cases}$$

Highlights of this work

- A simple model allowing for explicit calculations

some analytic results as a function of molecular spin S and/or lead polarization P

- Results are not trivial (there is some interesting physics in it)

spin blockade features, colossal spin fluctuations

- A perfect arena for testing different methods (comparison of usefulness and their consistency)

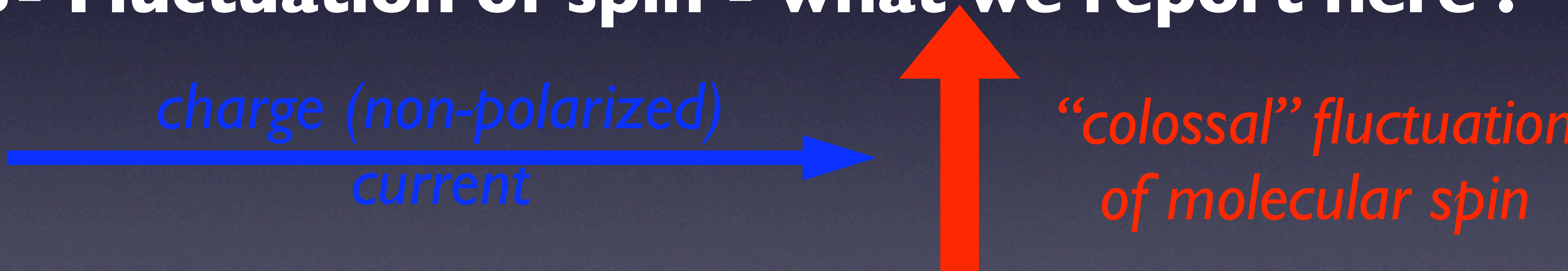
segment picture vs. FCS generating function

What do we attempt to “see” in this system?

1- Spin-dependent transport - effects of ferromagnetic electrodes

2- Manipulating a molecular spin - can we control the molecular spin by sending a (charge, spin) current?

3- Fluctuation of spin - what we report here :



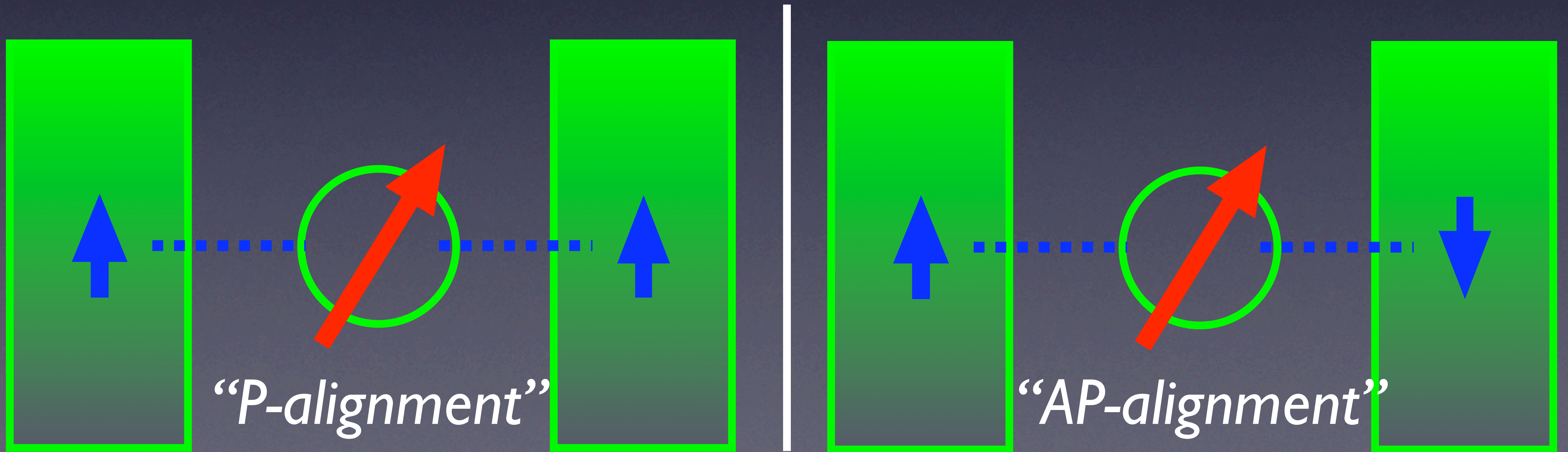
4- How to suppress such a huge enhancement of spin fluctuation?

In this work, we considered the cases of ...

1- Strong Coulomb blockade limit : $U \to \infty$
(double occupancy forbidden)
2- Incoherent tunneling regime - successive tunneling events are independent (cf. Master equation approach) justified at relatively high temperatures :

$$\hbar\Gamma \ll k_B T \ll eV$$

3- Collinear spin alignments of the electrodes :



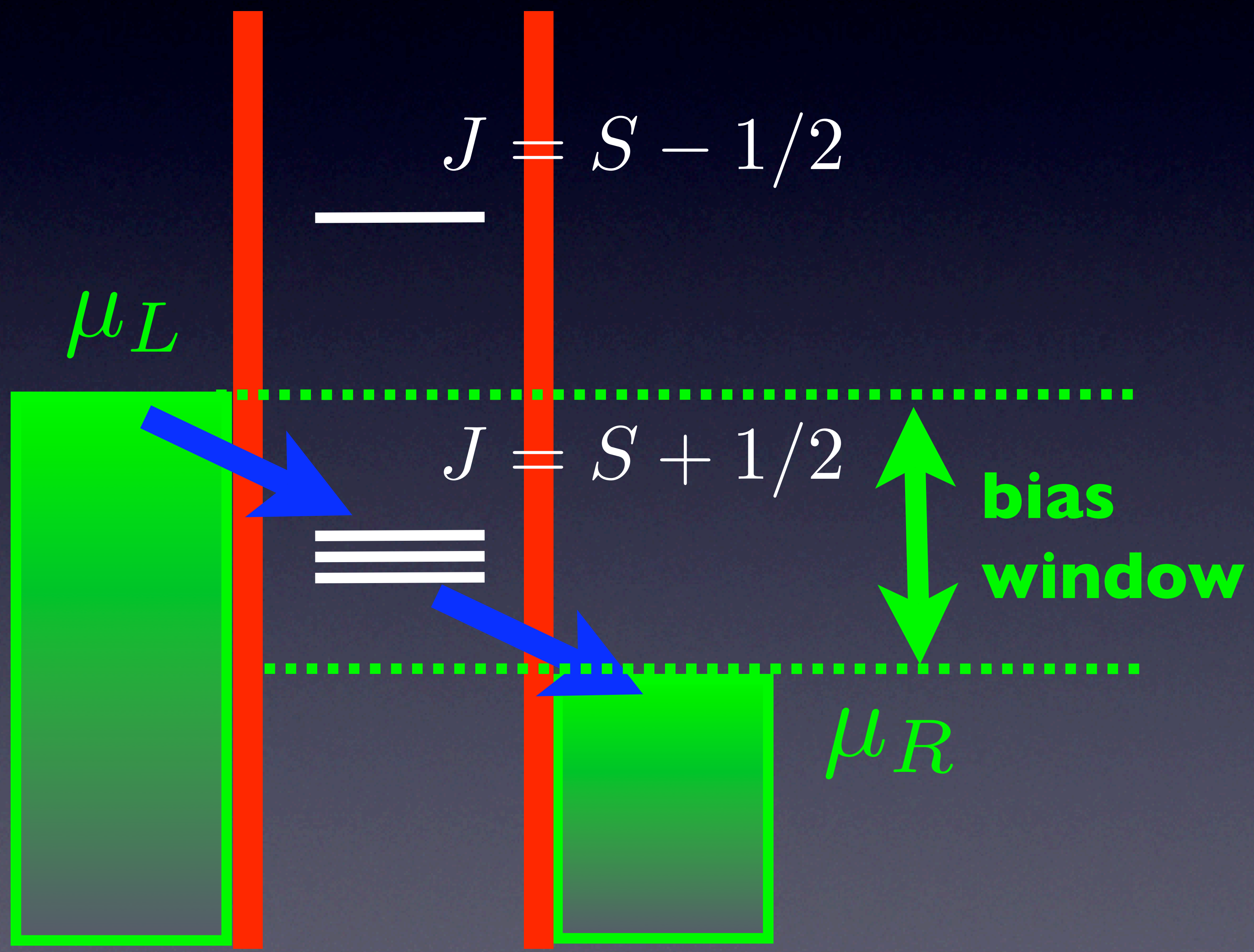
Even more specifically...

- a finite bias voltage

$$eV = \mu_L - \mu_R \gg k_B T$$

- electrons tunnel only from L to R

- ferromagnetic exchange interaction
- only J=S+1/2 spin sector in the bias window



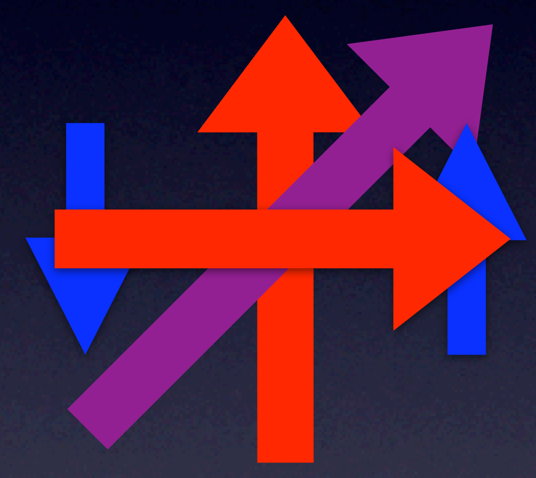
$J = S + 1/2$

$S_z = S$

$J_z = S - 1/2$

$\sigma_z = -1/2$

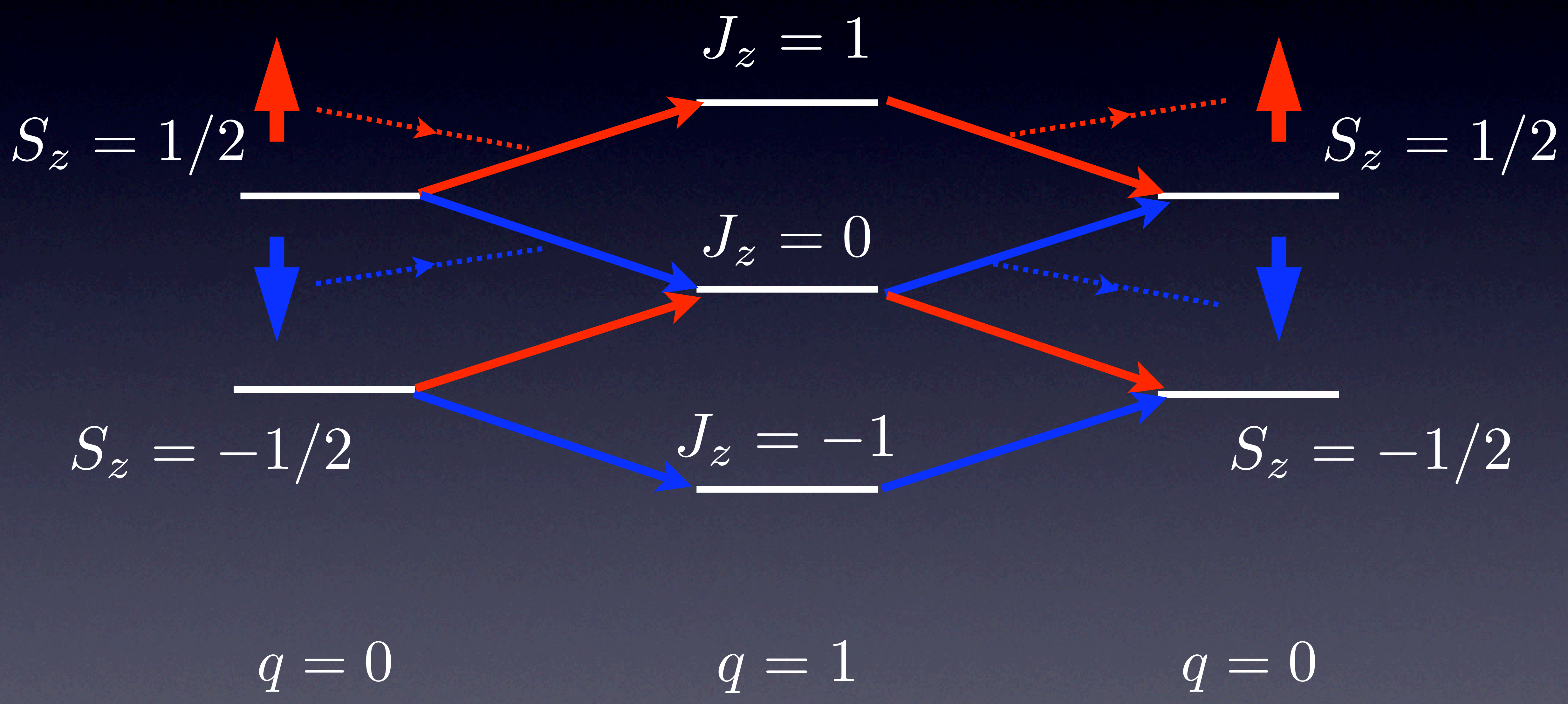
$\sigma_z = +1/2$



$S_z = S - 1$

S=1/2 case

only J=1 (triplet) spin sector in the bias window

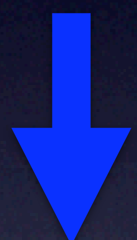


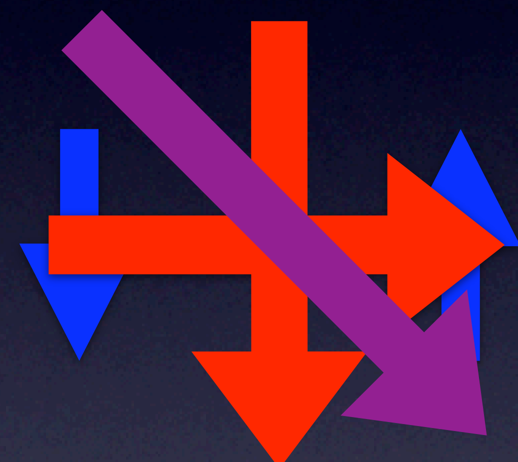
$$J = S + 1/2$$

$$S_z = S - 3/2$$

$$S_z = S - 1$$

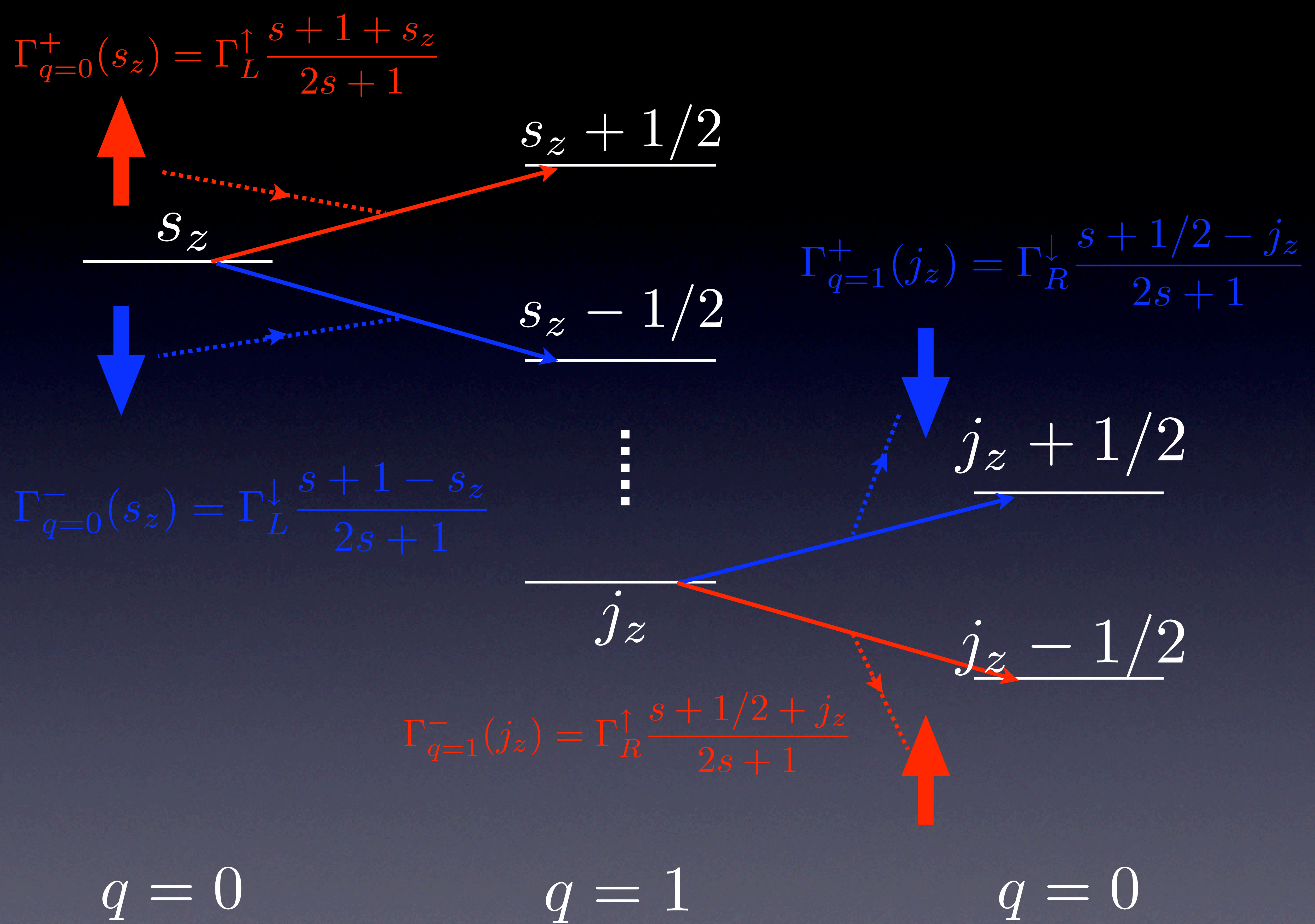
$\sigma_z = -1/2$





$\sigma_z = +1/2$

$$S_z = S - 2$$



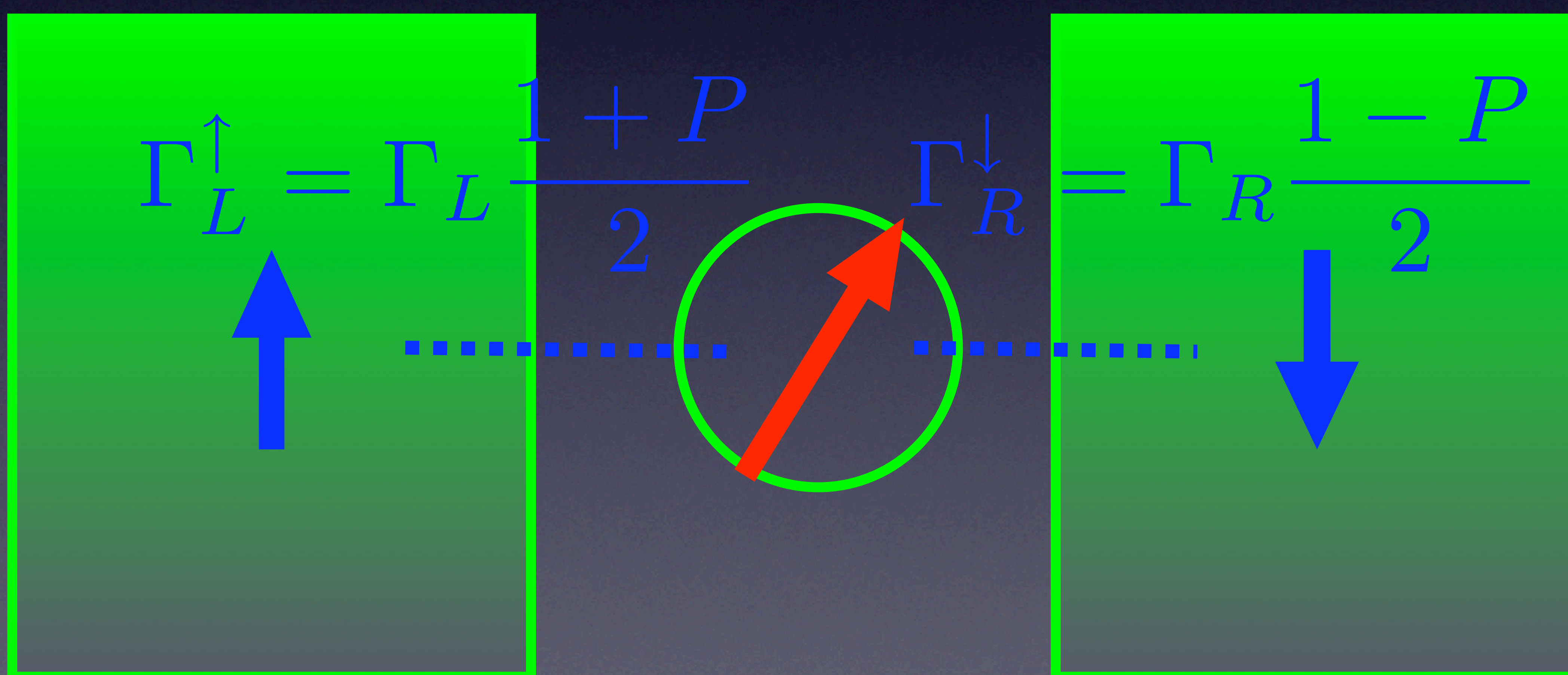
$\Gamma^{+}_{q=0}(s_z) = \Gamma^{\uparrow}_{L} \frac{s+1+s_z}{2s+1}$
$s_z + 1/2$
s_z
$s_z - 1/2$
$\Gamma^{+}_{q=1}(j_z) = \Gamma^{\downarrow}_{R} \frac{s+1/2-j_z}{2s+1}$
$j_z + 1/2$
$\Gamma^{-}_{q=0}(s_z) = \Gamma^{\downarrow}_{L} \frac{s+1-s_z}{2s+1}$
j_z
$j_z - 1/2$
$\Gamma^{-}_{q=1}(j_z) = \Gamma^{\uparrow}_{R} \frac{s+1/2+j_z}{2s+1}$
$q = 0$
$q = 1$
$q = 0$

What do we attempt to “see” in this system?

What are statistical averages of

(i) current *I*   (ii) charge Q   (ii) spin J_z

and their fluctuations as a function of P?



Actually, as far as current and charge are concerned, and also for $P=0$, we know "all" the correlation functions!

The idea of full counting statistics (FCS)

KI, Utsumi, Martin, PRB '07

The FCS generating function (exact solution) :

$$\Omega(\xi,\eta) = -\frac{z\Gamma_L + \Gamma_R - \xi}{2} + \frac{1}{2}\sqrt{(z\Gamma_L - \Gamma_R + \xi)^2 + 4z\Gamma_L\Gamma_R e^{\eta}}$$

$$z = \frac{2j+1}{2s+1}$$

The charge correlation functions :

$$\begin{aligned} \langle Q \rangle &= \left.\frac{\partial\Omega(\xi,\eta)}{\partial\xi}\right|_{\xi,\eta\to 0} = \frac{z\Gamma_L}{z\Gamma_L + \Gamma_R} \\ \langle QQ \rangle &= \left.\frac{\partial^2\Omega(\xi,\eta)}{\partial^2\xi}\right|_{\xi,\eta\to 0} = \frac{2z\Gamma_L\Gamma_R}{(z\Gamma_L + \Gamma_R)^3} \end{aligned}$$

The current correlation functions :

S : arbitrary
P=0

$$\begin{aligned} \langle I \rangle &= \left.\frac{\partial\Omega(\xi,\eta)}{\partial\eta}\right|_{\xi,\eta\to 0} = \frac{z\Gamma_L\Gamma_R}{z\Gamma_L + \Gamma_R} \\ \langle II \rangle &= \left.\frac{\partial^2\Omega(\xi,\eta)}{\partial^2\eta}\right|_{\xi,\eta\to 0} = \frac{z\Gamma_L\Gamma_R(z^2\Gamma_L^2 + \Gamma_R^2)}{(z\Gamma_L + \Gamma_R)^3} \end{aligned}$$

The segment picture

cf. Korotkov PRB '94

Time evolution of the dot state = a series of random jumps from one state to another

$$\cdots \to \alpha_0 \to \alpha_1 \to \alpha_2 \to \cdots \to \alpha_{M-1} \to \alpha_0 \to \cdots$$

a segment

$$|\alpha\rangle = |Q, J, J^z\rangle$$

We are interested in such quantities as

$$\int_0^T dt J_z(t) \to \sum_{n=1}^{N} \mathcal{J}_z[\xi_n], \quad \mathcal{J}_z[\xi] = \sum_{m=0}^{M-1} J_m^z \tau_m$$

Statistical average is done in two steps :

1- Poissonian average for a given segment

2- average over different realization of the segments

$$\begin{aligned} S_{J_z J_z} &= \frac{1}{T}\sum_{n=1}^{N}\left\langle \left[\mathcal{J}_z[\xi_n] - \frac{\langle \mathcal{J}_z\rangle}{\langle\tau\rangle}\tau[\xi_n]\right]^2\right\rangle \\ &= \frac{1}{\langle\tau\rangle}\left[\langle \mathcal{J}_z^2\rangle + \langle\tau^2\rangle\left(\frac{\langle \mathcal{J}_z\rangle}{\langle\tau\rangle}\right)^2 - 2\langle \mathcal{J}_z\tau\rangle\frac{\langle \mathcal{J}_z\rangle}{\langle\tau\rangle}\right] \end{aligned}$$

Application to molecular quantum dot magnet
(*s*=1/2 case : for comparison, *j*=1, *z*=3/2)

- **For P-alignment**

$$S_{QQ} = \frac{1}{\langle\tau\rangle}\left[\langle Q^2\rangle + \langle\tau^2\rangle\left(\frac{\langle Q\rangle}{\langle\tau\rangle}\right)^2 - 2\langle Q\tau\rangle\frac{\langle Q\rangle}{\langle\tau\rangle}\right] = \frac{24\Gamma_L\Gamma_R}{(1-p^2)(3\Gamma_L+2\Gamma_R)^3}$$

$$S_{J_zJ_z} = \frac{2(6\Gamma_L^2+4\Gamma_L\Gamma_R+\Gamma_R^2)}{(1-p^2)\Gamma_L\Gamma_R(3\Gamma_L+2\Gamma_R)}$$

- **For AP-alignment**

$$S_{QQ} = \frac{8(1-p^2)(3+32p^2+38p^4+40p^6+15p^8)\Gamma_L\Gamma_R}{\{(3+10p^2+3p^4)\Gamma_L+2(1-p^4)\Gamma_R\}^3}$$

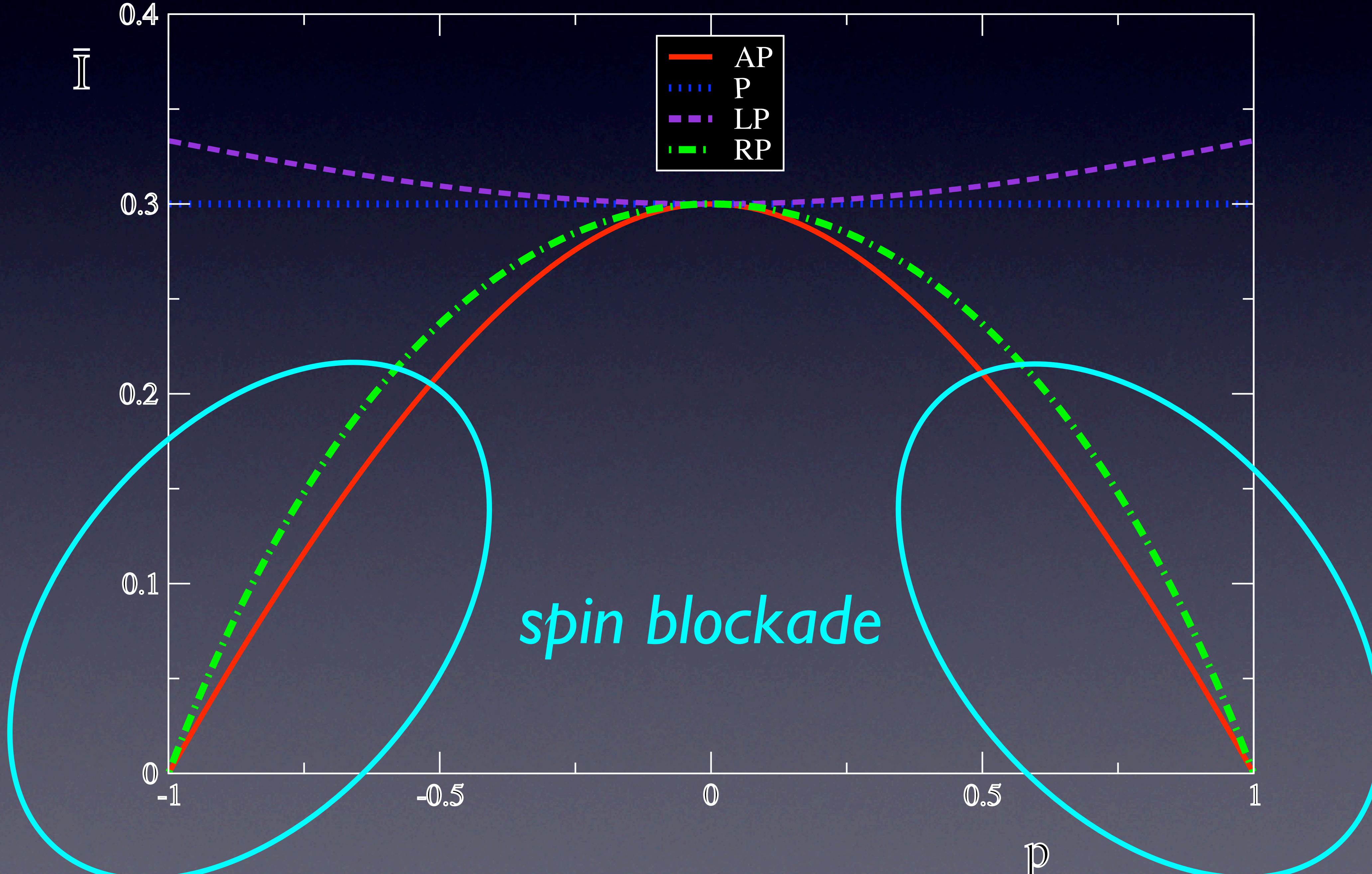
$$S_{J_zJ_z} = \frac{2(1-p^2)(275-200p^2+30p^4+48p^6-25p^8)}{(5+10p^2+p^4)^3}$$

Analytic formulae for the average current, charge and spin and their fluctuations (2nd and higher order noise correlations)

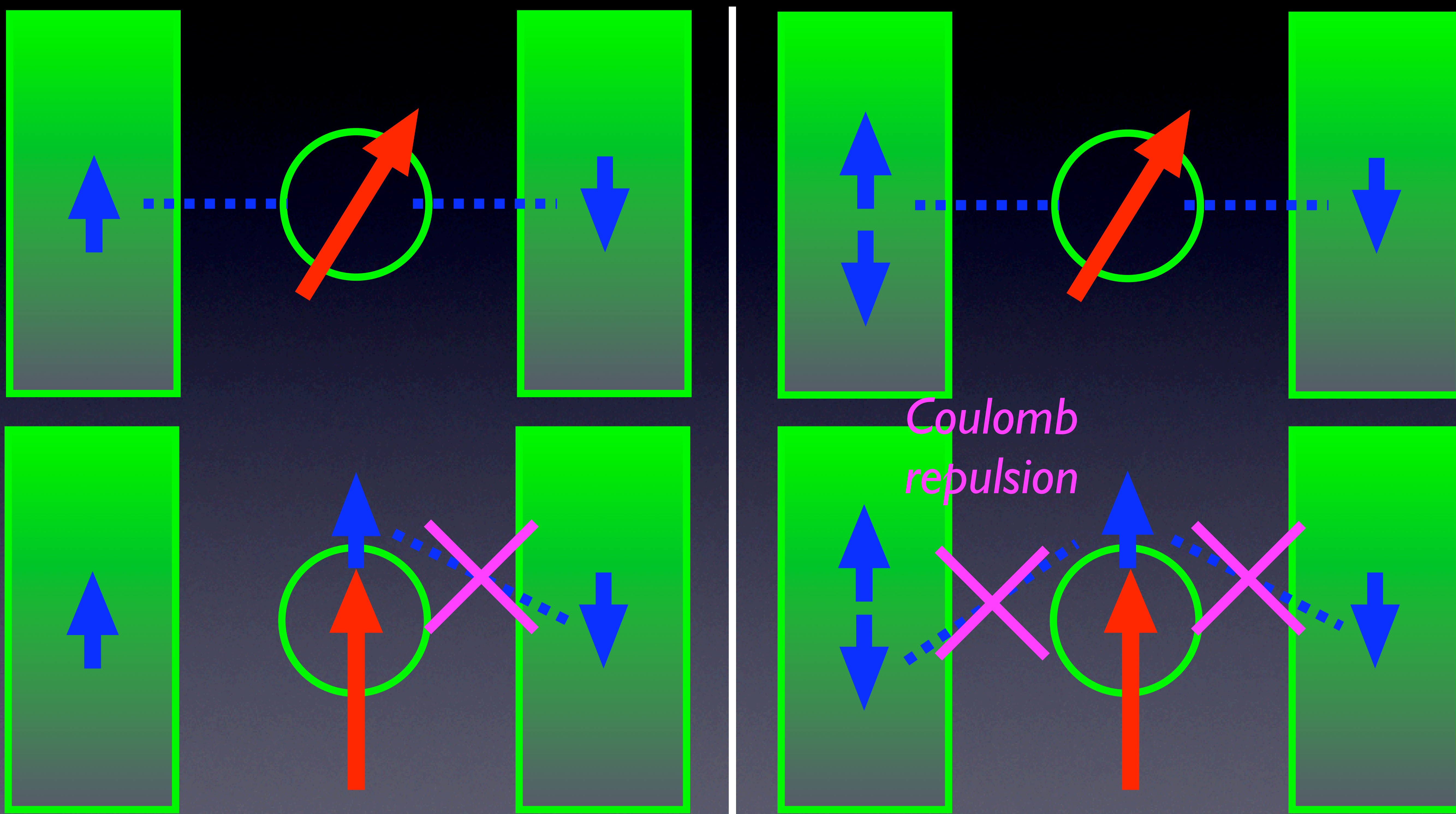
Spin blockade feature

Average current

- for a molecular spin s=1/2
- for ferromagnetic electrodes with various spin alignments



Spin blockade mechanism
- two slightly different patterns

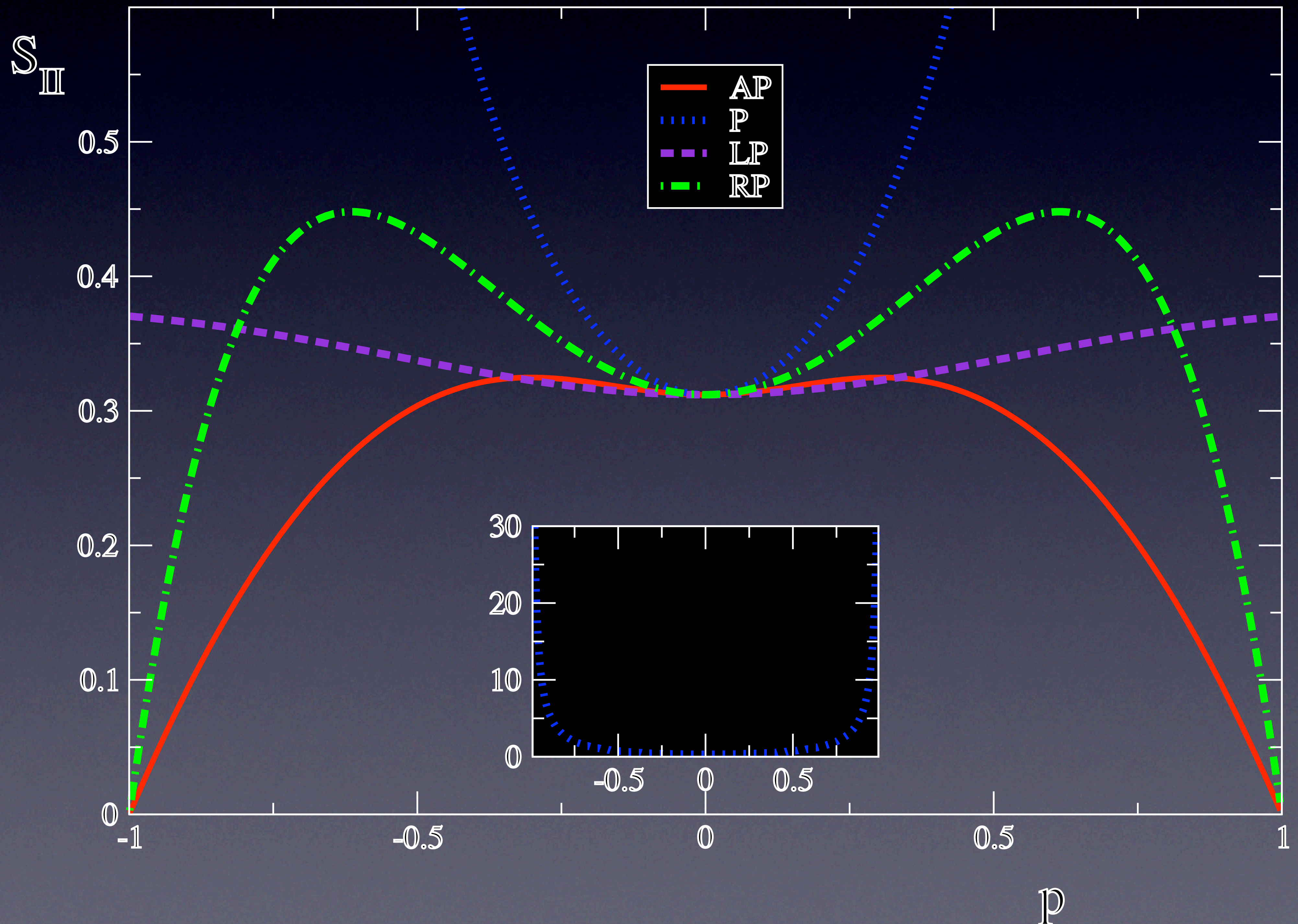


"AP"-alignment

"RP"- only Right electrode Polarized

Current fluctuation

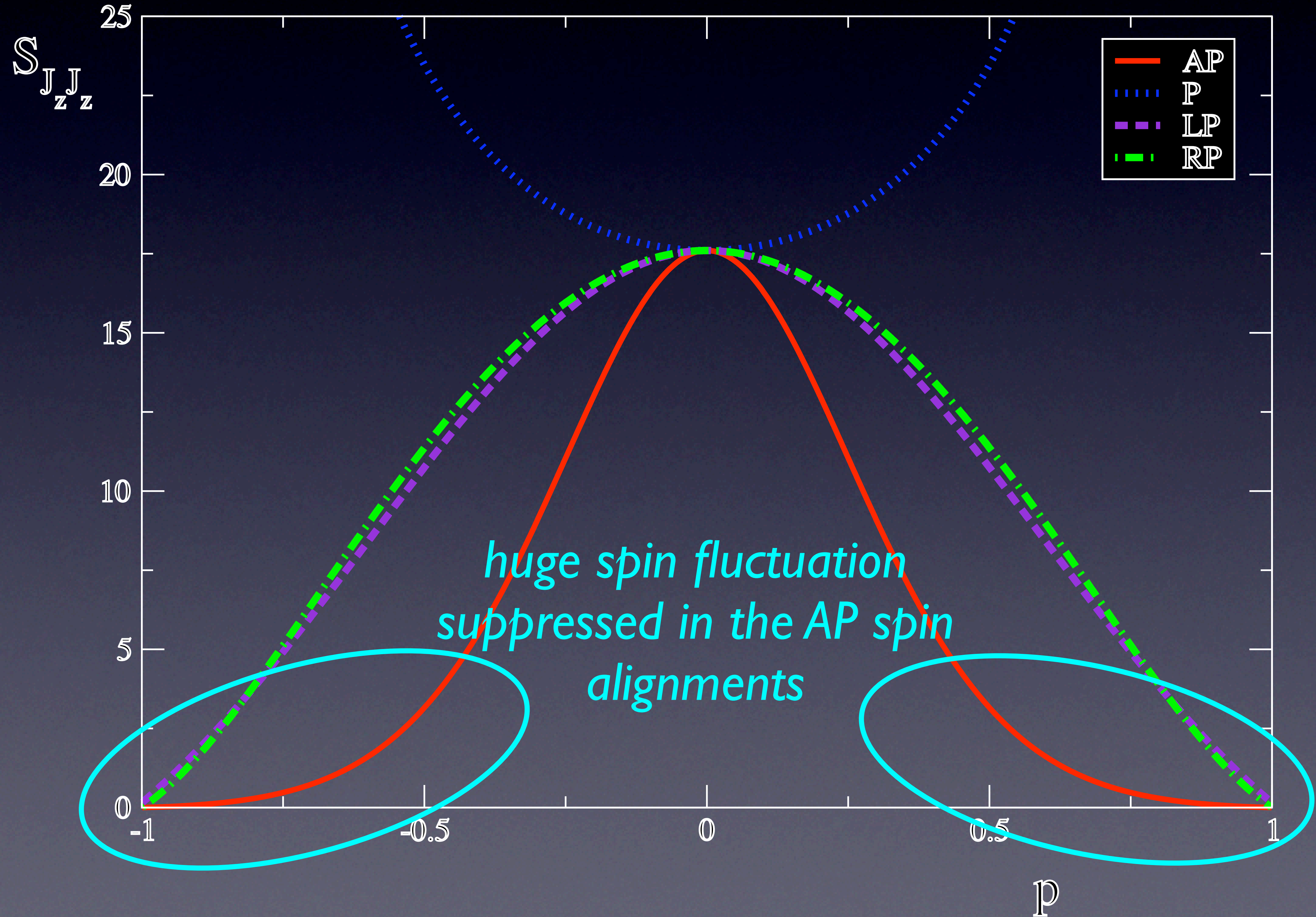
- for a molecular spin s=1/2
- for ferromagnetic electrodes with various spin alignments



“Colossal” spin fluctuation

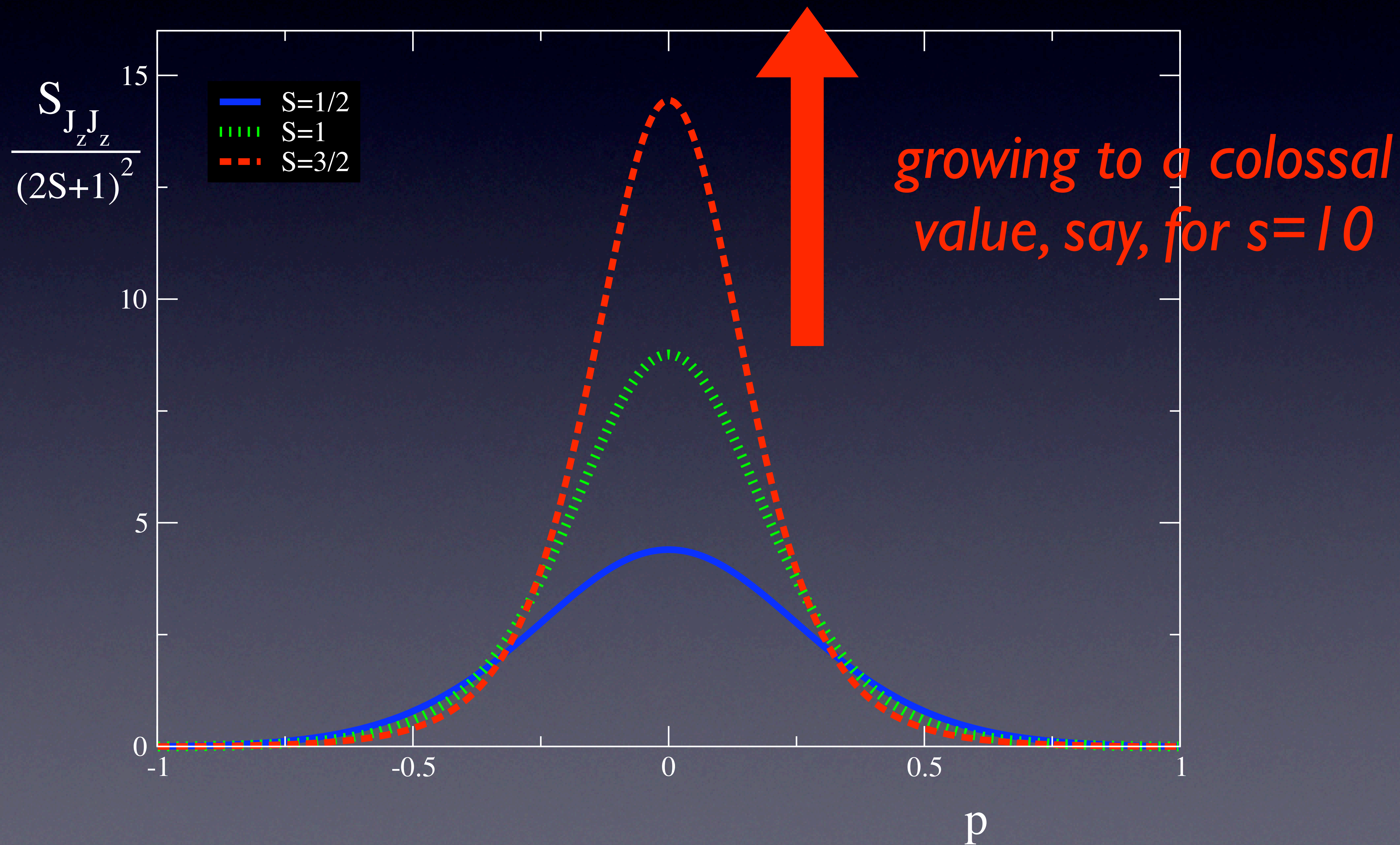
Spin fluctuation

- for a molecular spin s=1/2
- for ferromagnetic electrodes with various spin alignments



Spin fluctuation

- for a molecular spin s=1/2, 1, 3/2
- for ferromagnetic electrodes with AP alignments
- normalized by $(2s+1)^2$



Conclusions

1- Spin-dependent transport : ferromagnetic electrodes with *collinear* **spin alignments**

2- *Analytic formulae* **for the average current, charge and spin and their fluctuations (2nd and higher order noise correlations)**

3- Some specific features of spin transport, e.g., *spin blockade*, **etc. due to ferromagnetic leads**

4- Consistency with the *FCS method* **(for current and charge fluctuations and for** ***P=0*****)**

5- Colossal spin fluctuation : *possibly suppressed by ferromagnetic electrodes with AP spin alignments*

For further details, Jonckheere, KI, Martin, in preparation.