FirstPage

Magnetic QED, theoretical considerations.

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Formulas, a collection.

CONTENTS

I. INTRODUCTION

We develop the theory of magnetic QED.

II. ZEEMAN TERM (FIRST PRINCIPLES)

Our "fundamental theory" is the Pauli Hamiltonian (the non-relativistic limit of the Dirac equation). The magnetic field-spin coupling is through the Zeeman term. For a single electron it is given by [1, Chapter 2]:

$$H_{\mathrm{Z}} = -\frac{e\hbar}{2m}\vec{\sigma}\cdot\vec{B} = -\mu_B\vec{S}\cdot\vec{B} \tag{1}$$ HZ0

Here $\vec{\sigma}$ are the Pauli matrices (as given in Wikipedia, without 1/2). A more angular momentum way of writing is:

$$H_Z = -\frac{g_e\,\mu_B}{\hbar}\vec{s}\cdot\vec{B} \tag{2}$$ HZ1

here $\vec{s} = \frac{\hbar}{2}\,\vec{\sigma}$ (with eigenvalues $\pm\hbar/2$). Obviously $g_e = 2$. Some relevant constants and related quantities values are given in the table.

Constant	Value	Units (SI)
$\hbar$	1.05×10^{-34}	[J s]
μ_0	$4\pi\times10^{-7}$	$[\mathrm{NA}^{-2} = \mathrm{H/m} = \frac{\mathrm{J}}{\mathrm{mA}^2}]$
μ_B	9.27×10^{-24}	$[\mathrm{JT}^{-1} = \mathrm{J}\frac{\mathrm{kg}}{\mathrm{s}^2\mathrm{A}} = \mathrm{J}\frac{\mathrm{Kg}}{\mathrm{C\,s}}]$
$\frac{\mu_B^2\,\mu_0}{2\hbar}$	5×10^{-19}	$[\frac{\mathrm{m}^3}{\mathrm{s}}]$
ϵ_0	8.85×10^{-12}	$[\frac{\mathrm{C}^2}{\mathrm{Nm}^2}]$

Table I. Some constants and their units table:const

III. QUANTUM THEORY FOR THE CAVITY FIELD

sect:cav

We discuss the circuits and their quantum fluctuations. First, we remind the general formulas for the electromagnetic (EM) field. See [2, Chapt. 10] for a good and accessible first approach to the EM-quantization. See [3] for a complete quantization treatment. The EM energy is given by (through this manuscript SI-units are used):

$$E_{\text{EM}} = \frac{1}{2}\int dV\ \epsilon_0 E^2 + \frac{B^2}{\mu_0}\ . \tag{3}$$ Eem0

It is convenient to work with the vector potential $\vec{A}$. The magnetic and electrical field follow from it:

$$\vec{E} = -\frac{\partial \vec{A}}{\partial t} \tag{4}$$ EMax

$$\vec{B} = \vec{\nabla}\times\vec{A} \tag{5}$$ BMax

To solve for $\vec{A}$ and its quantization, we try the separation *ansatz*

$$\vec{A} = \sum_n \frac{1}{\sqrt{\epsilon_0 V_n}} q_n(t)\ \vec{u}_n(\vec{r})\ . \tag{6}$$ sep

Here, $\vec{u}_n$ **is dimensionless and** $[q_n] = [\sqrt{\hbar/\omega}] = [\sqrt{\text{Js}^2}]$. It can be checked that $[A] = \text{Jsm}^{-1}/\text{C}$.

The dimensioless $\vec{u}_n$ are basis vectors (normal modes for the EM-field)

$$\frac{1}{\sqrt{V_n V_m}}\int dV \vec{u}_n\cdot\vec{u}_m = \delta_{nm}\ , \tag{7}$$

and

$$\frac{1}{\sqrt{V_n V_m}}\int dV\ \vec{\nabla}\times u_n\cdot\vec{\nabla}\times\vec{u}_m = k_n^2\delta_{nm}\ , \tag{8}$$

with V_n the n-mode volume. Therefore, the energy (3) is given by:

$$E_{\text{EM}} = \frac{1}{2}\sum_n \dot{q}_n^2 + \omega_n^2 q_n^2\ . \tag{9}$$ Eem

A. The CPW case (single mode)

We now **quantize** the EM-field for the CPW [Cf. Fig. 1]. We do it within the **single mode approximation**. In the **Coulomb gauge**,

$$q = \sqrt{\frac{\hbar}{2\omega_c}}(a^\dagger + a) \tag{10}$$ qq

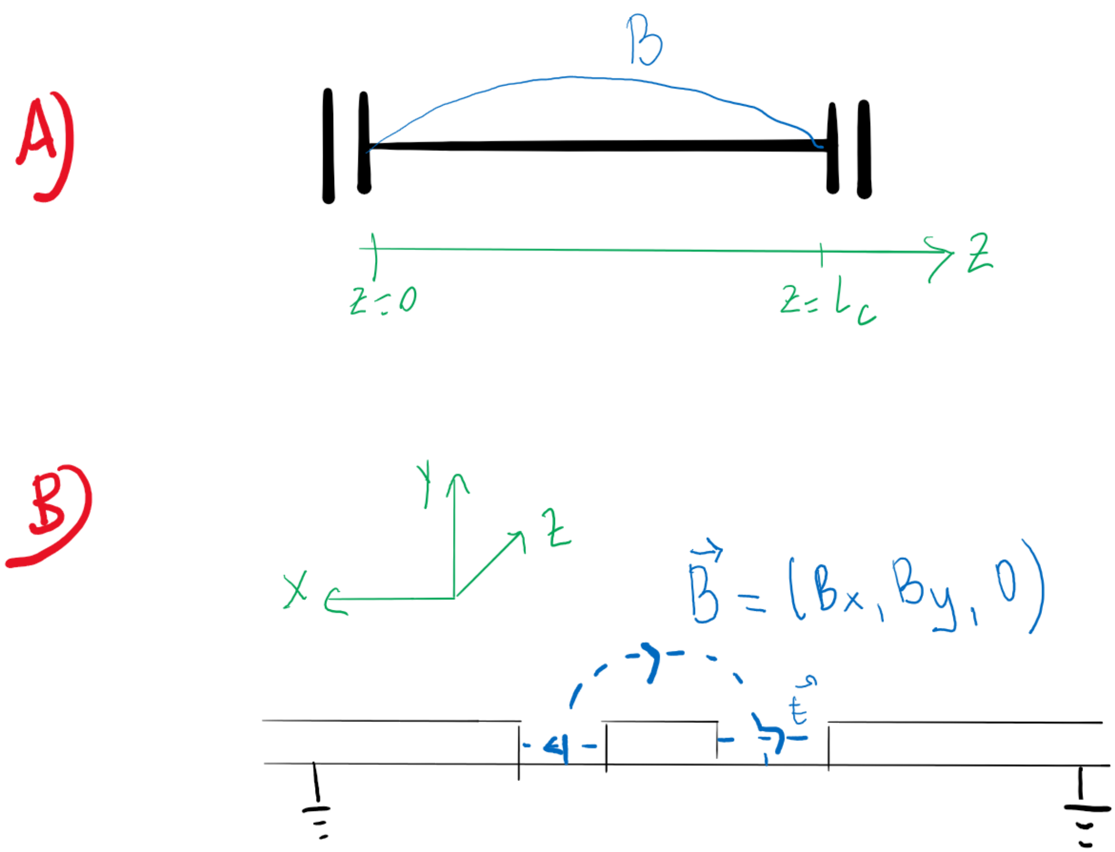

Figure 1. a) Top view of a resonator (length $= L_c$). The z dependence of the field amplitude is shown for the $\lambda/2$-mode. b) cut of the resonator. The coordinates are shown. The field is within the xy-plane. fig:cpw

It is easy to guess the corresponding energy operator. From (9) and (10), $\hat{E}_{\mathrm{EM}} = \frac{1}{2}\sum_n p_n^2 + \omega_n^2 q_n^2$. We will not use the $\hat{}$ anymore. From now on, everything is quantum. Besides, the quantum vacuum energy fluctuation is: $\langle 0|E_{\mathrm{EM}}|0\rangle = \frac{1}{2}\hbar\omega_c$. In particular, from the virial theorem we know that magnetic and electric energy is

$$\frac{1}{2}\int dV\ \epsilon_0 \langle 0|E^2|0\rangle = \frac{1}{2}\int dV\ \frac{\langle 0|B^2|0\rangle}{\mu_0} = \frac{1}{4}\hbar\omega_c \tag{11}$$ viral

This will be very important in the following.

For the CPW [See fig. 1] and with the single mode approximation,we have that

$$\vec{u} = u(x,y)\frac{1}{\sqrt{2}}\sin(kz)\mathbf{z}\ , \tag{12}$$ uxyz

with $k = \pi/L_c$, L_c is the cavity length.

Putting altogether the magnetic field generated in the CPW reads [Cf. Eqs. (5), (6), (10) and (12)],

$$\begin{aligned}\vec{B} &= \sqrt{\frac{c^2 \hbar \mu_0}{4V\omega_c}}\, \sin(kz)\big(\partial_y u(x,y)\mathbf{x} - \partial_x u(x,y)\mathbf{y}\big)\left(a^\dagger + a\right) \\ &\equiv \vec{B}_{\rm rms}(\vec{r})(a^\dagger + a) \end{aligned} \tag{13}$$

Bcav

here, $\vec{B}_{\rm rms}$ is the root-mean-square magnetic field fluctuations.

Finally, noticing that

$$\langle 0|B^2|0\rangle = B^2_{\rm rms} \tag{14}$$

and using (11) we get that:

$$\frac{1}{2}\int dV \frac{B^2_{\rm rms}}{\mu_0} = \frac{1}{4}\hbar\omega_c \tag{15}$$

Brmshwc

This is another important result.

1. *For the calculation of* $\vec{B}_{\rm rms}$

We want to provide a recipe for calculating $\vec{B}_{\rm rms}$ with finite element numerical tools. For that, we must *inject* some current, that we call $I_{\rm rms}$. To calculate the latter, we rewrite the EM energy in terms of current and voltage :

$$E_{\rm EM} = \frac{1}{2}\Big(C_{\rm eff} V^2 + L_{\rm eff} I^2\Big)\ . \tag{16}$$

EEMVI

Here, $C_{\rm eff}$ ($L_{\rm eff}$) are the effective capacitance (inductance) for the CPW. The effective inductance is given by:

$$L = \frac{2Z}{\pi\omega_c} \tag{17}$$

Leff

Here,

$$Z = \sqrt{\frac{l}{c}} \tag{18}$$

the Transmission line impedance c (l) are the capacitance (inductance) per unit of length. Besides the CPW frequency is given by ($\lambda/2$-mode)

$$\omega_c = vk = \frac{1}{\sqrt{lc}}\frac{\pi}{L_c} \tag{19}$$

Using (11) and (15), we obtain:

$$\frac{1}{2}\int dV \frac{B_{\rm rms}^2}{\mu_0} = \frac{L I_{\rm rms}^2}{2} = \frac{1}{4}\hbar\omega_c \to I_{\rm rms}^2 = \frac{\hbar\omega_c}{2L} = \frac{\hbar\pi\omega_c^2}{4Z} \quad (20) \quad \boxed{\texttt{Irms}}$$

In the last equality we have used (17).

B. LC-resonator

If we consider a LC-resonator instead, Eq. (16) is (of course) valid. On the other hance, for the LC-resonator we know that $\omega_{\rm LC} = \frac{1}{\sqrt{LC}}$ ($Z_{\rm LC} = \sqrt{L/C}$ Therefore we obtain for the $I_{\rm rms}$ [Cf. Eq. (20) and (17)],

$$I_{\rm rms}^2 = \frac{\hbar\omega_{\rm LC}^2}{2Z} \quad (21)$$

IV. SPIN(S)-CIRCUIT COUPLING

sect:s-c

We have the tools to estimate the single spin - single photon coupling. Also, the coupling to an ensemble. Using Eqs. (1) or (2) and (13), the coupling Hamiltonian reads,

$$H = -\frac{g_e\mu_B}{\hbar}\vec{s}\cdot\vec{B}_{\rm rms}(a^\dagger + a)\ . \quad (22) \quad \boxed{\texttt{gspincavity}}$$

A. Single spin and xy-plane field

If we particularize to the case of $s = 1/2$ and magnetic field in the XY-plane we can simplify it:

$$H_{s=1/2} = -\frac{g_e}{2}\mu_B|B_{\rm rms}|\left(e^{i\theta}\sigma^+ + {\rm h.c}\right)(a + {\rm h.c.})\ , \quad (23) \quad \boxed{\texttt{Hs1/2}}$$

with $B_{{\rm rms},x} + iB_{{\rm rms},y} = |B_{\rm rms}|e^{i\theta}$. Finally, the coupling is given by:

$$\boxed{\hbar g = \frac{g_e}{2}\mu_B|B_{\rm rms}| \qquad \text{single spin } (s = 1/2) \text{ coupling}} \quad (24) \quad \boxed{\texttt{gss}}$$

B. Coupling to an ensemble

If we consider a $s = 1/2$-spin ensemble instead we generalize Eq. (23) to:

$$H_{\rm s=1/2\ ensemble} = \sum_j g_j(e^{i\theta_j}\sigma_j^+ + {\rm h.c.})(a^\dagger + a)\ . \quad (25)$$

It is useful to define the collective operator

$$b^\dagger = \frac{1}{\sqrt{N}} \frac{1}{\sqrt{\overline{g^2}}} \sum_j g_j e^{i\theta_j} \sigma_j^\dagger \tag{26}$$ bdef

with

$$\overline{g^2} = \sum_j \frac{g_j^2}{N} \tag{27}$$

Notice that this is the correct definition because the bosonic relation $[b, b^\dagger] \cong 1$ is obtained in the low-polarization limit [4]. Using the identification (26) we obtain the resonator-ensemble coupling:

$$H_{s=1/2 \text{ ensemble}} = \hbar g_{\text{eff}} (b + b^\dagger)(a + a^\dagger) \tag{28}$$

with

$$\boxed{g_{\text{eff}} = \sqrt{N}\sqrt{\overline{g^2}} \qquad \text{ensemble } (s = 1/2) \text{ coupling}} \tag{29}$$ gse

C. Some estimations

We give some estimations that may be useful for comparison with the numerical calculations or for understanding the experiments.

1. *Ensemble (large)*

Consider the case of a big sample, big enough to cover the entire *coupling region*, *i.e.* the sample cover the whole mode volume. If we assume constant density:

$$g_{\text{eff}} = \sqrt{N}\sqrt{\overline{g^2}} \to \sqrt{\rho \int g^2 dV} = \sqrt{\rho}\, \frac{g_e}{2} \sqrt{\frac{\mu_B^2\, \mu_0\, \omega_c}{2\hbar}} \tag{30}$$ geffbound1

In the continuum limit we have used that $\sum_j f_j = \frac{1}{\Delta V} \sum_j \Delta V f_j \to \rho \int f \, dV$, since $\Delta V = V_0/N = \rho^{-1}$. V_0 is the mode-volume (we use the suffix 0 to emphasize that we are dealing with the cavity fundamental mode). In the last equality we have used (15). Using the results of table I we get (set $g_e = 2$):

$$g_{\text{eff}} = \sqrt{5 \times 10^{-19} [\text{m}^3/\text{s}]\ \rho\, \omega_c} \tag{31}$$ geffbound2

This is a rather simple formula that bounds the achievable coupling.

Obviously, a smaller ensemble (not covering the whole mode volume) gives less coupling than (30) and (31).

2. Single spin and/or small ensemble

We take the naivest approach. We assume that the position dependence for the field ($B_{\rm rms}$) can be computed within the Biot-Lavart law and considering an infinite wire, namely:

$$B_{\rm rms}(x, y, z = L_c/2) = \frac{\mu_0}{2\pi r} I_{\rm rms}(z = L_c/2) \; . \tag{32}$$

We have fixed the spins to be at the optimal $z = L_c/2$ point. Here $r = \sqrt{x^2 + y^2}$ with the origin located at the middle of the central superconducting conductor. Therefore, using (24) and (20) and making that $I_{\rm rms}(z = L_c/2) = \frac{1}{\sqrt{2}} I_{\rm rms}$ we get:

$$g = \frac{\mu_B \mu_0}{\sqrt{\pi \hbar Z}} \frac{1}{4r} \omega_C \tag{33}$$

with $\frac{\mu_B \mu_0}{4\sqrt{4\hbar Z}} = 2.23 \times 10^{-14}$ m, where we have set $Z = 50$ Ohms. Setting, $r = w$ (the width of the central line) we can estimate the maximum coupling for a single spin. For example, *e.g.* for a $w = 0.01\mu$m we get the coupling $g \cong 2.23 \times 10^{-6} \omega_C$. Setting the cavity frequency $\omega_C \sim$ GHz we finally estimate a coupling of the order of KHz. This is in agreement with Mark numerical estimation (Fig 5 in the ACS nano draft).

A small ensemble scales as $\sqrt{N} g$ (if all the spins see the same field).

D. $S > 1/2$ and beyond CPW generalization

We generalise the coupling Hamiltonian to deal with $S > 1/2$ molecules. In general, the Zeeman coupling is:

$$H_Z = -\frac{g_e \mu_B}{\hbar} \vec{S} \cdot \vec{B} \tag{34}$$ Hzgral

Here $[S_i, S_j] = i\hbar \epsilon_{ijk} S_k$. Notice that $[S_i] = [\hbar]$.

Recall that quantization yields,

$$\vec{B} = \vec{B}_{\rm rms}(a^\dagger + a) \tag{35}$$

The magnetic field may have components in the three spatial directions $\vec{B}_{\rm rms}(\vec{r}) = (B_x, B_y, B_z) = |\vec{B}_{\rm rms}|(\sin\theta \, \cos\phi, \sin\theta \, \sin\phi, \cos\theta) = B_\perp \hat{u}_\perp + B_z \hat{k}$. Here, $B_\perp$ and B_z are the projections on the xy and z-plane respectively. It is convenient to introduce the ladder operators:

$$S_\pm = S_x \pm i S_y \; . \tag{36}$$ ladder

Then, (34) can be rewritten:

$$\begin{aligned} H_Z &= -\frac{g_e\mu_B}{\hbar}\vec{B}_{\mathrm{rms}}\cdot\vec{S}\left(a^\dagger + a\right) \\ &= -\frac{g_e\mu_B}{\hbar}\left(\frac{B_\perp}{2}\left(e^{i\phi}S_+ + \mathrm{h.c.}\right) + B_z S_z\right)\left(a^\dagger + a\right). \end{aligned} \tag{37}$$

HZladder

Wich are two ways of writing the Zeeman coupling.

Finally, imagine that we are interested in compute the coupling to two states. E.g. the states defining the qubit. I.e. we take two states from the $(2S+1)-$states of H_S. Let us denote these states $\{|0\rangle, |1\rangle\}$ (following the standard notation). We also define the operators:

$$\tau^+ = |1\rangle\langle 0| \tag{38}$$

$$\tau^- = |0\rangle\langle 1| \tag{39}$$

$$\tau_z = \sigma_z \tag{40}$$

wich they are nothing but the Pauli-related matrices (I use another notation to avoid confusion with the $S=1/2$ case, Here we do not have a $S=1/2$ but a larger spin from wich we select two levels). Then, we can write the zeeman term (restricted to these two states) as,

$$H_Z = \frac{g_e\mu_B}{\hbar}\left(\left(\langle 1|\vec{B}_{\mathrm{rms}}\cdot\vec{S}|0\rangle\,\tau^+ + \mathrm{h.c.}\right) + \Lambda_{z,-}\tau_z + \Lambda_{z,+}I_2\right)\left(a^\dagger + a\right) \tag{41}$$

HZ01

where we have defined

$$\Lambda_{z,\pm} \equiv \frac{1}{2}\Big(\langle 0|\vec{B}_{\mathrm{rms}}\cdot\vec{S}|0\rangle \pm \langle 1|\vec{B}_{\mathrm{rms}}\cdot\vec{S}|1\rangle\Big) \tag{42}$$

Here I_2 is the 2×2 identity matrix. This Hamiltonian can be rewritten as

$$H_z = \hbar\big((g_\perp e^{i\phi}\tau_+ + h.c) + g_z\tau_z\big)(a^\dagger + a) \tag{43}$$

where $g_\perp$ and g_z follow from (41) straightforwardly. The I_2 term yields a displacement in the cavity (trivial, I guess) so we did not consider. Finally, this last equation (23) in the $S=1/2$case and the CPW.

V. QUANTUM THEORY FOR THE TRANSMISSION LINE

sect:TL

If we consider the spin(s) coupled to a transmission line instead, we must change (a little) the Hamiltonian, See figure 2. Going back to the quantization discussion in Sect. III, we

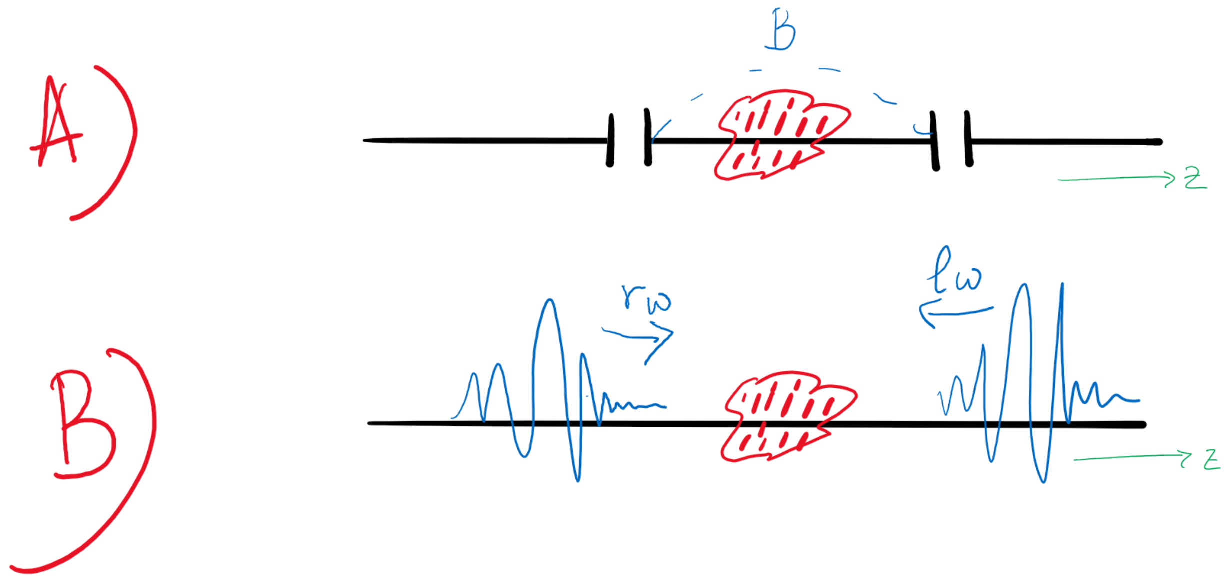

Figure 2. a) Top view of a resonator. The z dependence of the field amplitude is shown for the $\lambda/2$-mode. c) Top view of a TL the spins are coupled to both right and left moving photons. fig:tlcav

can write the magnetic field generated in the transmission line in a fully quantum way:

$$\vec{B}(\vec{r}) = \int d\omega \, \vec{\mathcal{B}}(\vec{r}, \omega) \, (r^\dagger_\omega + l^\dagger_\omega) + \text{h.c.} \; . \tag{44}$$

Please, notice that $[\mathcal{B}] = [B/\sqrt{\omega}]$. Therefore, the Zeeman coupling adds to the line and spin Hamiltonians as [Cf. with Sect. IV and Eq. (**??**)]:

$$\begin{aligned} H =& H_{\text{spins}} + H_{\text{line}} + H_{\text{I}} \\ =& H_{\text{spins}} + \hbar \int \mathrm{d}\omega \, \omega \, (r^\dagger_\omega r_\omega + l^\dagger_\omega l_\omega) + \hbar \left(e^{i\phi} \frac{S_+}{\hbar} + \text{h.c.} \right) \int \mathrm{d}\omega \, g(\omega) \, (r^\dagger_\omega + l^\dagger_\omega) + \text{h.c.} \end{aligned} \tag{45}$$ Htotal

In (45) we have distinguished between right (left) moving photons with the operators $r^\dagger_\omega$ ($l^\dagger_\omega$). Below, we find a relationship between the spectral density ($\equiv g(\omega)$) and the spin-cavity coupling, see next section V A. We have assumed a non chiral coupling, *i.e.* $g(\omega)$ is the same for for right and left moving photons.

To simplify the notation we define

$$S \equiv \left(e^{i\phi} \frac{S_+}{\hbar} + \text{h.c.} \right) \tag{46}$$ Sdef

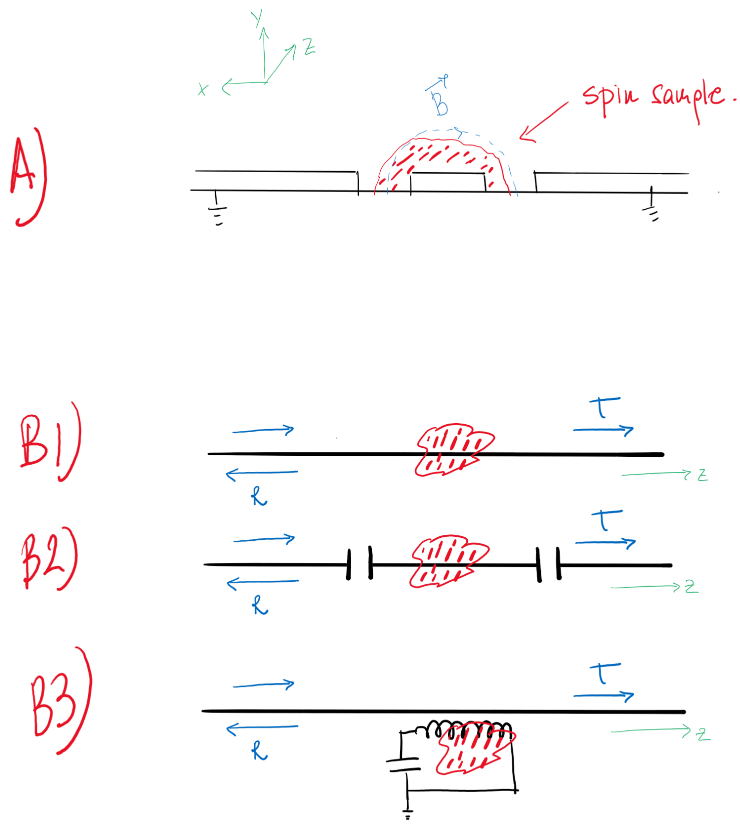

Figure 3. a) cut of a transmission lines (fixing the coordinates) b) Different situations to be discussed: b1) open TL, b2) transmission measurements for resonators, b2) LC-resonators side-coupled to open transmission lines. fig:t-situa

A. Relation between g and $g(\omega)$

ct:relation

There is a relation between $g(\omega)$ in Eq. (45) and g in (22) or (**??**). It is given by:

$$g = \sqrt{2 \times 2\pi g^2(\omega) \times \frac{\omega_c}{\pi}} \tag{47}$$ g-gw

TO DO: David must write the the derivation of (47).

VI. TRANSMISSION (THEORY)

sec:trans

Formulas for the transmission (reflection) and how to compute them in practice are briefly summarized. Three situations are of interest here. They describe the experiments of spins coupled to either open transmission lines, CPW or resonators, See Fig. 3.

A. Input-output theory.

sec:inout

Transmission and reflection formulas are based on the input-output theory, wich relates the current injected (input field) and the transmitted or reflected (output field). Input-output formalism is nothing but scattering theory [5]. Formally, the in and out fields relation is by means of the Heisenberg evolution for the right and left photon operators. After some algebra it is known that

$$r_\omega(t_f)\,\mathrm{e}^{i\omega t_f} = r_\omega(t_0)\,\mathrm{e}^{i\omega t_0} - i\sum_n g_n(\omega)\int_{t_0}^{t_f} \mathrm{d}\tau \mathrm{e}^{i\omega\tau} S_n(\tau)\;. \tag{48}$$ iod0

and exactly the same for the left ones,

$$l_\omega(t_f)\,\mathrm{e}^{i\omega t_f} = l_\omega(t_0)\,\mathrm{e}^{i\omega t_0} - i\sum_n g_n(\omega)\int_{t_0}^{t_f} \mathrm{d}\tau \mathrm{e}^{i\omega\tau} S_n(\tau)\;. \tag{49}$$ iod0l

t_0 is the initial time. In scattering, t_0 marks the time in which the incident wave packet is injected in the line. The final time, t_f, is any time after the interaction between the wavepacket and the scatterers occurred.

Now, input and output fields are defined as

$$r_{\mathrm{in}}(t) := \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}}\, r_\omega(t_0)\mathrm{e}^{-i\omega(t-t_0)} \qquad l_{\mathrm{in}}(t) := \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}}\, l_\omega(t_0)\mathrm{e}^{-i\omega(t-t_0)} \tag{50}$$ ain

and

$$r_{\mathrm{out}}(t) := \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}} r_\omega(t_f)\mathrm{e}^{-i\omega(t-t_f)} \qquad l_{\mathrm{out}}(t) := \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}} l_\omega(t_f)\mathrm{e}^{-i\omega(t-t_f)}\,. \tag{51}$$ aout

Within the above definitions we rewrite (48) as,

$$r_{\mathrm{out}}(t) = r_{\mathrm{in}}(t) - i\sum_n \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}}\int_{t_0}^{t_f} \mathrm{d}\tau\; g_n(\omega)\mathrm{e}^{-i\omega(t-\tau)} S_n(\tau) \tag{52}$$ io0

Now, making $t_0 \to -\infty$ and $t_f \to \infty$, (52) yields:

$$r_{\mathrm{out}}(t) = r_{\mathrm{in}}(t) - i\sum_n \int_0^\infty \frac{\mathrm{d}\omega}{\sqrt{2\pi}} \mathrm{e}^{-i\omega\, t} g_n(\omega)\int_{-\infty}^{\infty} \mathrm{d}\tau\; \mathrm{e}^{+i\omega\tau} S_n(\tau) \tag{53}$$ io1

and identical relation for the left fields. This is a central result. It links the input and output fields through the dynamics of the system operator dynamics $S(\tau)$, cf. Eq. (46). Obviously, in this section we have considered the situation of different spins coupled (n-sum).

B. Initial condition

Experiments are performed at finite temperature. Bot the line and spins are initially thermalized at temperature T. The *inject* current is a coherent state generated over the thermal state. Thus, this input field can be written as,

$$\varrho_{\rm in} = \mathcal{A}\,\varrho_\beta\,\mathcal{A}^\dagger \tag{54}$$

with $\mathcal{A}$ the generator of coherent states,

$$\mathcal{A} = \sum_k \phi_k \mathrm{e}^{\alpha_k a_k^\dagger - \alpha_k^* a_k} \tag{55}$$ Acoh

C. Transmission, Reflection formulas

First, we deal with the situation specified in Fig. 3a), *i.e.* spins coupled to an open line. The transmission $t(\omega)$ is given by,

$$t(\omega) = \frac{\langle \alpha_\omega | r_{\rm out}(t) | \alpha_\omega \rangle}{\langle \alpha_\omega | r_{\rm in}(t) | \alpha_\omega \rangle} \tag{56}$$ t-c

with

$$|\alpha_\omega\rangle = \mathrm{e}^{\alpha r_{\rm in}^\dagger(\omega) - \mathrm{H.c.}} |\Omega\rangle \tag{57}$$ alpha

being $r_{\rm in}^\dagger(\omega)$ the Fourier transform of $r_{\rm in}^\dagger(t)$.

We are going to calculate the transmittance in the linear regime (i.e. the transmission does not depend on the intensity) [Cf. Eq. (55)]:

$$t(\omega) = \lim_{\alpha \to 0} \frac{\langle \alpha_\omega | r_{\rm out}(t) | \alpha_\omega \rangle}{\langle \alpha_\omega | r_{\rm in}(t) | \alpha_\omega \rangle} \tag{58}$$ t1-c

The reflection is,

$$r(\omega) = t(\omega) - 1 \tag{59}$$ rt1

D. Single Spin calculation

Let us consider the case of a single spin. We will come to the total transmission latter on. To obtain the transmission, Eqs. (56), (58) and (52), we need

$$\langle S(t) \rangle = \mathrm{Tr}(S \varrho)\;. \tag{60}$$ St

In principle, the model (45) can not be solved exactly. Therefore, we need to do several approximations. All of them are well justified. First, we assume that the spin-line coupling can be treated perturbatively. Then, tracing out the waveguide we end up with the Born-Markov master equation.

$$\begin{aligned}\frac{d\varrho}{dt} =& - i[H_{\rm spins}, \varrho] - i\, \alpha g(\omega) 2\cos(\omega t)[S(t), \varrho] \\ &+ 2\sum_{\omega_{ij}} \Gamma_{ij}(\omega_{ij})\Big(\hat{P}_{ij}\varrho\hat{P}^\dagger_{ij} - \frac{1}{2}\{\hat{P}^\dagger_{ij}\hat{P}_{ij}, \varrho\}\Big) + \mathcal{D}\varrho\mathcal{D} - \frac{1}{2}\{\mathcal{D}^2, \varrho\}\,,\end{aligned} \tag{61}$$ qme-0

Here, α is the amplitude of the input current, Cf. Eqs. (55) and (57). In the master equation it acts as an external driving of frequency ω. $\hat{P}_{ij} = |i\rangle\langle j|$ are relaxation operators connecting allowed transitions (as induced by $\hat{S}$) between the $H_{\rm spins}$ eigenstates $|i\rangle$ and $|j\rangle$. The thermal rates are:

$$\begin{aligned}\Gamma_{ij}(\omega_{ij} > 0) &= \left(2\pi g^2(\omega_{ij})|\langle i|S|j\rangle|^2 + \widetilde{\gamma}_{ij}\right)\left(n_{\omega_{ij}} + 1\right) \equiv \mathcal{G}_{ij} + \gamma_{ij} \\ \Gamma_{ij}(\omega_{ij} < 0) &= \left(2\pi g^2(\omega_{ij})|\langle i|S|j\rangle|^2 + \widetilde{\gamma}_{ij}\right)\left(n_{\omega_{ij}}\right) = e^{-\beta\omega_{ji}}\Gamma_{ij}(\omega_{ji})\end{aligned} \tag{62}$$ Gammas

Here $n_{\omega_{ij}} = \left(e^{\beta\omega_{ij}} - 1\right)^{-1}$ is the bosonic occupation number. The last equality in the second line is detailed balance which ensures thermalization. Notice that, in addition to the spontaneous emission into the transmission line [See $\mathcal{G}$ in (62)] we also consider the *intrinsic* losses of the sample: $\widetilde{\gamma}_{ij}(n_{\omega_{ij}} + 1) = \gamma_{ij}$ wich in nothing but T_1^{-1} of the resolved transition $i \leftrightarrow j$. Finally $\mathcal{D}$ a diagonal matrix accounting for pure dephasing. Finally, the 2 in from of the sum is because the master equation is coupled both to left (l_ω) and right (r_ω) modes.

$\langle S\rangle$ is the response of the scatterer to the input current (see the driving term in (61)). The transmission experiments that we want to describe do not depend on the driving amplitude. In other words, we are within the linear response theory regime. Then,

$$\langle S(\tau)\rangle = S(\omega){\rm e}^{i\omega_0\tau} + S^*(\omega){\rm e}^{-i\omega_0\tau} \tag{63}$$

Replacing the latter in (53) we get,

$$\begin{aligned}\langle r_{\rm out}(t)\rangle &= \langle r_{\rm in}(t)\rangle - i\int_0^\infty \frac{{\rm d}\omega}{\sqrt{2\pi}}{\rm e}^{-i\omega\, t} g(\omega)\int_{-\infty}^{\infty} {\rm d}\tau\ {\rm e}^{+i(\omega-\omega_0)\tau}S(\omega) + {\rm e}^{+i(\omega+\omega_0)\tau}S^*(\omega) \\ &= \langle r_{\rm in}(t)\rangle - i\sqrt{2\pi}g(\omega_0)S(\omega_0){\rm e}^{-i\omega_0 t}\,,\end{aligned} \tag{64}$$ io-lrt

Therefore, using (58) we finally arrive to:

$$t^{(1)}(\omega) = 1 - i2\pi g(\omega_0)\frac{1}{\alpha}S(\omega_0) \tag{65}$$ tside

and

$$r^{(1)}(\omega) = -i2\pi g(\omega_0)\frac{1}{\alpha}S(\omega_0) \tag{66}$$

rside

Using the definition of susceptibility, $S(\omega) = \alpha g(\omega)\chi_{\hat{S},\hat{S}}$ we obtain the transmission as a function of the latter:

$$t^{(1)}(\omega) = 1 - i2\pi g(\omega)^2 \chi_{\hat{S},\hat{S}}(\omega) \tag{67}$$

rside

Recall that $\chi_{\hat{S},\hat{S}}(\omega)$ is the usual notation for the susceptibility for $\hat{S}$ when the spin ensemble is driven via $\hat{S}$, see the driving term in (61).

Using (B4) we obtain the *single spin* transmission.

$$t^{(1}(\omega) = 1 - \sum_{\omega_{ij}=\omega} \frac{\mathcal{G}_{ij}\Delta P_{ij}}{\mathcal{G}_{ij} + \gamma_{ij} + i(\omega_{ij} - \omega)} \tag{68}$$

Here,

$$\Delta P_{ij} \equiv \frac{e^{-\beta\epsilon_j} - e^{-\beta\epsilon_i}}{\mathcal{Z}_0} \tag{69}$$

A final approximation is considering only the two levels closest to resonance with the incident photons. Therefore, there is no sum over i and j. Writing $\Delta E = \epsilon_i - \epsilon_j$ (the transiton frequency).

$$t^{(1)} = 1 - \frac{\mathcal{G}\Delta P}{\mathcal{G} + \gamma + i(\Delta E - \omega)} \tag{70}$$

E. Total transmission (open line)

Let us consider now the total transmission through N-spins. Using the transfer matrix we know that:

$$t = \frac{1}{1 - \sum_n r_n^{(1)}/t_n^{(1)}} \tag{71}$$

ttotal

Using (59) we obtain,

$$\frac{r_n^{(1)}}{t_n^{(1)}} = -\frac{\mathcal{G}_n\Delta P}{\mathcal{G}_n(1-\Delta P) + \gamma + i(\Delta E - \omega)} \cong -\frac{\mathcal{G}_n\Delta P}{\gamma + i(\Delta E_n - \omega)} \, . \tag{72}$$

rt

We have assumed that the spontaneous emission in the line can different for each spin ($\mathcal{G}_n$) because each spin is at different position respect the line. However, we assume that the intrinsic losses are the same for every spin. Eqs. ((71)) and ((72)) together with the identification

$$\gamma = T_1^{-1} + T_2^{-1} \tag{73}$$

is the formula for the total transition.

TO DO: David must write the the derivation of (47).

F. Units and dependence

Notice that $[g(\omega)] = \sqrt{\omega}$, See (45) and (47). Therefore $[g^2(\omega)] = [\mathcal{G}] = \omega$. This is expected since the spontaneous emission is proportional to $\mathcal{G}$. Finally, $\mathcal{G}$ is a function of the frequency. For one dimensional lines $\mathcal{G}\omega$. Therefore, we can always write $\mathcal{G} = \lambda\omega$, with λ a dimensionless constant.

VII. RESONATOR PLUS MOLECULES HAMILTONIAN

sec:HcQED

A. Spins coupled to a CPW resonator

Now, we compute the transmission when the spins are coupled to a CPW cavity. In this case, the spin ensemble and the cavity are the system under study (the one in which the spectroscopy is done). The CPW is coupled to two semi infinite lines, see. Fig. 3b2.

The resonator-ensemble Hamiltonian is [Cf. Eq. (**??**) and (46)]:

$$H = H_{\rm spin} + \hbar\omega_c a^\dagger a + \hbar \sum_n g_n S_n (a + a^\dagger) \; . \tag{74}$$ QED-single

The CPW - transmission line coupling is characterized by a photon leackage onto the latter at a given rate $\sim \kappa$. This may be a fitting parameter. Recall that $\kappa \sim 1/Q$ with Q the resonator quality factor.

Following Appendix B we know that:

$$t(\omega) = -i \frac{1}{\alpha} \langle a \rangle \tag{75}$$

Here $\langle a \rangle$ is the response [Cf. with (65)] The e.o.m. for $\langle a \rangle$ with external driving is:

$$0 = -i(\Omega - \omega)\langle a \rangle - i\alpha\kappa - i \sum_n g_n \langle \hat{S}_n \rangle - \kappa \langle a \rangle \; . \tag{76}$$

Now, each $\hat{S}_n$ induces a transition between two levels. We can, then use the algebra of the Pauli matrices . Noticing that $\langle \hat{S}_n \rangle \to \langle \sigma_n^- \rangle = {\rm Tr}(\varrho \sigma^-) = \varrho_{10}$, then

$$0 = -i(\Delta E - \omega)\langle \sigma_n^- \rangle - \gamma \langle \sigma_n^- \rangle + i g_n \langle a \rangle \Delta P \tag{77}$$

Putting all together we end up with

$$t(\omega) = \frac{-\kappa}{(\Omega - \omega) - i\kappa + i \frac{\Delta P \sum g_n^2}{i(\Delta E - \omega) + \gamma}} \tag{78}$$ tres

Using (47) we get that

$$g_n = \sqrt{\frac{2\mathcal{G}_n(T=0)\omega_c}{\pi}} \tag{79}$$

B. Spins coupled to a lumped element resonator

Instead of a CPW resonator we can consider a lumped element resonator (LER), which is side coupled to the transmission line (see Fig. 3b3). The hamiltonian and e.o.m. from the CPW resonator are still valid. However, for the LER-spins system the transmission is given by equation (65) (see Appendix B):

$$t(\omega) = 1 - \frac{\kappa}{(\omega_c - \omega) - i\kappa + i\frac{\Delta P \sum g_n^2}{i(\Delta E - \omega) + \gamma}} \tag{80}$$ tres_LE

C. Fitting formulas

Rewriting equation 78 as,

$$t(\omega) = \frac{\kappa}{(\omega - \widetilde{\omega}_c) + i\widetilde{\kappa}} \tag{81}$$

$$\widetilde{\kappa} = \kappa + \frac{\gamma}{(\Delta E - \omega)^2 + \gamma^2} \Delta P \sum g_n^2 \tag{82}$$

$$\widetilde{\omega}_c = \omega_c + \frac{\omega - \Delta E}{(\Delta E - \omega)^2 + \gamma^2} \Delta P \sum g_n^2 \tag{83}$$

These formulas coincide (we are in resonant conditions $\Delta E \cong \Omega$) with the ones used in Mark thesis.

Appendix A: Side versus interrupted

sec:side

Notice that this resembles related experiments with an open line. In that case, there is not resonator, but just the spins placed close the TL. In the latter, the system is the spin ensemble and it is not interrupting the line but placed close to it. We can denote this situation as a system *side coupled* to the line (in contrast to the resonator case). By taking a look to the figure 4 we can understand that interrupted and side coupled are complementary. In fact (up to a phase)

$$t_i = r_s \quad \text{and } r_i = t_s \tag{A1}$$

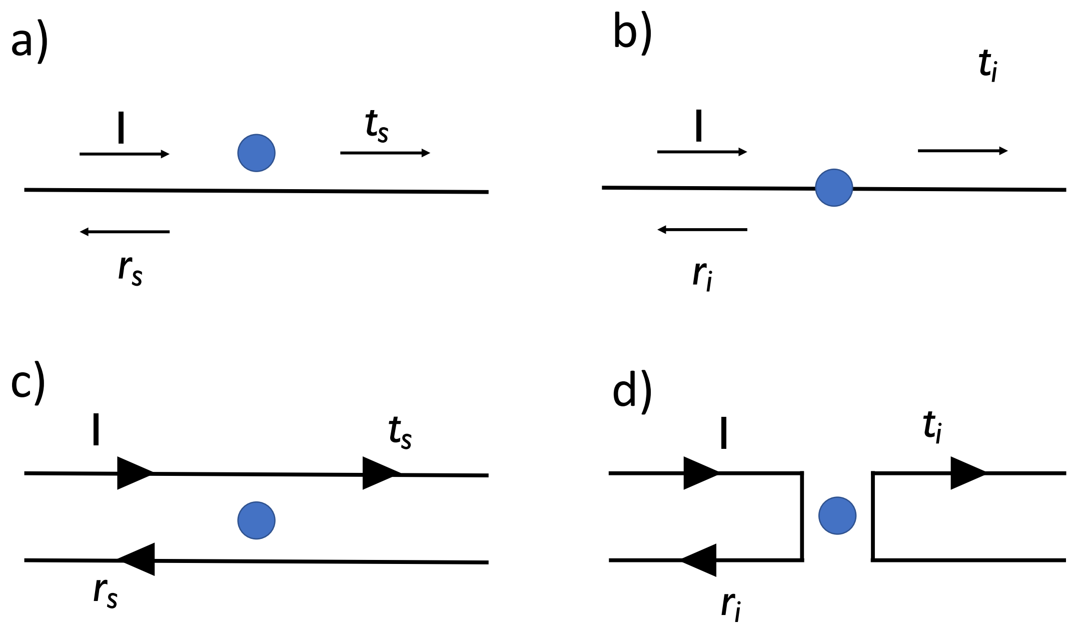

Figure 4. (Color online) a) Side coupled b) interrupted case c) left-right moving photons for the side case d) equivalent view for the interrupted.

fig:svsi

where r_i (r_s) stands for the reflection in the interrupted (side coupled) case and equivalent notation for the transmission coefficients. Therefore, we can use the formalism done for open lines in this case. This is really great.

Appendix B: Calculation of the dynamical susceptibility

sec:side

In the system eigenbasis the time evolution for the off-diagonal matrix elements is

$$\varrho_{ij} = \varrho_{ij}(0)e^{(-i\omega_{ij}+\Gamma_{ij})t} \tag{B1}$$

Now, notice that

$$\langle S \rangle = \sum_{ij} A_{ij}\varrho_{ji}(t) \tag{B2}$$

In addition $\varrho_{ij}(0)$ can be computed perturbatively (with pertrurbation $+\alpha g(\omega)S$):

$$\varrho_{ji}(0) = \alpha g(\omega)\frac{\langle j|\hat{S}|i\rangle}{\omega_{ji}}\frac{e^{-\beta\epsilon_j} - e^{-\beta\epsilon_i}}{\mathcal{Z}_0} \tag{B3}$$

Here ϵ_i are the system energies and $\omega_{ij} = \epsilon_i - \epsilon_j$. With this we end up

$$\begin{aligned}\chi_{\hat{S},\hat{S}} &= \sum_{\omega_{ji}\cong\omega} \frac{|\langle i|\hat{S}|j\rangle|^2}{\omega_{ji}} \frac{e^{-\beta\epsilon_j} - e^{-\beta\epsilon_i}}{\mathcal{Z}_0} \frac{\Gamma_{ji} + i\omega_{ji}}{\Gamma_{ji} + i(\omega_{ji} - \omega)} \\ &\cong -i \sum_{\omega_{ji}\cong\omega} |\langle i|\hat{S}|j\rangle|^2 \frac{e^{-\beta\epsilon_i} - e^{-\beta\epsilon_j}}{\mathcal{Z}_0} \frac{1}{\Gamma_{ji} + i(\omega_{ji} - \omega)}\end{aligned} \tag{B4}$$ chiSS-app

which is the expression used in the main text by exchanging $i \leftrightarrow j$

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