Theory of a type of quantum amplification: Phase-sensitive amplification by frequency upconversion

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It is shown that the spectroscopic bridge, a recently proposed method to achieve high-efficiency microwave oscillators and phase-sensitive detectors, may be used in a spin multiplet of certain paramagnetic ions to obtain a type of microwave quantum amplification: phase-sensitive amplification by frequency upconversion. This operation of a solid-state maser without population inversion is based on a principle similar to that used for achieving amplification in a transistor (triode). The major differences between this amplification and the parametric amplification, or the usual double-resonance maser action, are discussed.

The definition of a linear amplification includes both frequency-converting amplification and phase-sensitive amplification.¹ In usual single- or multiple-resonance experiments,² a high signal-to-noise ratio arises from phase-sensitive detection with respect to the microwave or radio-frequency field, and with respect to an applied Zeeman modulation field. However, the modulation frequency and the relaxation rates must not be comparable, so this frequency is much lower than that of the detected signal.

Low noise amplification is provided in the maser devices³ and parametric amplifiers,⁴ which are both (except for the degenerate paramp) phase-insensitive amplifiers.¹ Unfortunately, in the usual case of double-resonance maser action, the better amplification is achieved at very low temperatures. To maintain such low temperatures entails, on one hand, considerable expense and inconvenience, and, on the other hand, a weak emitting field. When this field is strong, the nonlinear and saturating effects become important. These limitations, together with very rapid developments in the area of parametric amplifiers, have resulted in a virtual cessation of development in the solid-state maser field.

Unlike the usual maser action, in the case of triple resonance, when the spectroscopic bridge conditions are fulfilled, high-efficiency microwave generations or phasesensitive detections have recently been predicted.⁵ Although all three microwave fields are strong, important linear or nonsaturating effects occur, while the heat absorbed by the lattice per unit time is minimum.

The purpose of the present work is to show that within the spectroscopic-bridge (SB) method, the operation of a solid-state maser without inversion based on a principle similar to that used in a transistor (triode) is a type of microwave quantum amplification that combines the above amplification methods, and at the same time is a phasesensitive amplification.

Consider a nondegenerate multilevel spin system in a dilute paramagnetic solid, with unequally spaced energy levels and whose simple transitions are well separated:

$$\omega_{ii}^{o} - \omega_{rs}^{o} | \gg (T_2^{ij})^{-1}, \ i \neq r$$
, (1)

where $\omega_{ij}^{o} = (E_i - E_j) / \hbar$, E_i being the eigenvalues of the

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spin Hamiltonian, and $(T_2^{ij})^{-1}$ is the linewidth of a homogeneously broadening simple transition. Let us consider three levels n, σ, m among the levels mentioned above: $E_n > E_{\sigma} > E_m$, and three quasimonochromatic fields at resonance,⁵ whose frequencies are correlated: $\omega_{nm} \cong \omega_{n\sigma} + \omega_{\sigma m}$, where $\omega_{ij} = \omega_{ij}^o + \delta \omega_{ij}$ and where this correlation condition is fulfilled as long as

$$\delta\omega_{ij} \ll (T_2^{ij})^{-1} . \tag{2}$$

In this case, we should place our sample into a dc magnetic field and a cavity capable of supporting these three orthogonal fields.

Let us suppose that all these fields are strong enough so that

$$\langle p_{ij}^2 \rangle - \langle p_{rs}^2 \rangle \ll \langle p_{ij}^2 \rangle \gg (T_2^{ij})^{-1} (T_2^{rs})^{-1}$$
, (3)

where p_{ij} are matrix elements (written in the interaction representation and expressed in \hbar unit) of the Hamiltonian, which represents the interactions of the multilevel spin system with the fields mentioned above. In this case, the heat absorbed by the lattice per unit time: $P_L = -(P_{nm} + P_{n\sigma} + P_{\sigma m})$ is minimum and independent on the field intensities. On the contrary, the powers P_{ij} absorbed or emitted by the sample depend strongly on these intensities. Let it be some small intensity changes, $d \langle p_{ij}^2 \rangle \ll \langle p_{ij}^2 \rangle$. As a consequence of the remark mentioned above, $dP_{nm} + dP_{n\sigma} + dP_{\sigma m} \approx 0$. The corresponding power changes are $dP_{ij} = 2(E_i - E_j)N d\Omega_{ij}$, where N is the total number of spins, while the parameters Ω_{ij} are defined in Ref. 5. As the three frequencies are correlated, $d\Omega_{ij}$ have the following property:

$$-d\Omega_{nm} \simeq d\Omega_{n\sigma} \simeq d\Omega_{\sigma m} \simeq d\Omega = \delta\Omega_{mn} + \delta\Omega_{n\sigma} + \delta\Omega_{\sigma m} ,$$
(4)

where

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$$\begin{split} \delta\Omega_{n\sigma} &= [(\rho_n^{\sigma} - \rho_{\sigma}^{o})(T_2^{mn}T_2^{n\sigma} + T_2^{mn}T_2^{\sigma m} + T_2^{n\sigma}T_2^{\sigma m} \\ &+ 2T_2^{mn}T_{nm}^{mn} + 2T_2^{\sigma m}T_{m\sigma}^{\sigma m}) \\ &+ (\rho_{\sigma}^{o} - \rho_m^{o})2T_2^{\sigma m}T_{\sigma m}^{n\sigma} \\ &+ (\rho_m^{o} - \rho_n^{o})2T_2^{mn}T_{mn}^{n\sigma}]\Gamma^{-1}d\langle p_{n\sigma}^2 \rangle , \end{split}$$

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$$\begin{split} \Gamma &= [T_{2}^{mn} T_{2}^{n\sigma} + T_{2}^{mn} T_{2}^{\sigma m} + T_{2}^{n\sigma} T_{2}^{\sigma m} \\ &+ 2T_{2}^{mn} T_{nm}^{mn} + 2T_{2}^{\sigma m} T_{m\sigma}^{\sigma m} \\ &+ 2T_{2}^{n\sigma} T_{\sigma n}^{n\sigma} + 4(T_{\sigma n}^{n\sigma} T_{m\sigma}^{\sigma m} - T_{n\sigma}^{\sigma m} T_{\sigma m}^{n\sigma})] \\ &\times [(T_{2}^{mn})^{-1} + (T_{2}^{n\sigma})^{-1} + (T_{2}^{\sigma m})^{-1}] \,. \end{split}$$

 $\delta\Omega_{mn}$ and $\delta\Omega_{\sigma m}$ are obtained by circular permutations of $mn, n\sigma$, and $\sigma m. \rho_i^o = N_i^o/N$ and N_i^o is the thermal equilibrium population of the level E_i . $T_{ij}^{jr} = T_{ji}^{rl} = K_{ij}^j - K_{ij}^r$ and $T_{ji}^{il} > T_{ij}^{ir} > 0$. The spin-lattice relaxation times K_{ij}^r are obtained by inverting the W matrix corresponding to the spin-lattice relaxation rates w_{ij} .⁵

As a consequence of the property (4), as long as the conditions (3) are fulfilled, the power change dP_{ij} corresponding to any small changes of the three-field intensities is proportional to the frequency ω_{ij} , while dP_{nm} has always opposite sign in comparison with $dP_{n\sigma}$ and $dP_{\sigma m}$ (see Fig. 1):

$$\frac{dP_{n\sigma}}{\omega_{n\sigma}} \cong \frac{dP_{\sigma m}}{\omega_{\sigma m}} \cong -\frac{dP_{nm}}{\omega_{nm}} \cong 2\hbar N \, d\Omega \, . \tag{5}$$

This important property could be used for a highefficiency phase-sensitive amplification by frequency upconversion. For instance, let us consider

$$(T_2^{n\sigma})^{-1} \ll \omega_{n\sigma} \ll \omega_{nm}, \omega_{\sigma m} \tag{6}$$

and the following spectroscopic-bridge conditions:

$$\langle p_{n\sigma}^{o2} \rangle = \langle p_{\sigma m}^{o2} \rangle , \qquad (7a)$$

$$\langle p_{nm}^{o2} \rangle = \langle p_{n\sigma}^{o2} \rangle - (T_2^{n\sigma})^{-1} (T_2^{\sigma m})^{-1} + p^2 ,$$
 (7b)

$$p^2 \ll \langle p_{ij}^{o2} \rangle . \tag{7c}$$

When the conditions (3) and (7) are fulfilled, the linear effects prevail and $P_{nm} < 0$, $P_{n\sigma} > 0$, and $P_{\sigma m} > 0$ (see Fig. 1). In other words, the microwave power P_{nm} is absorbed, while the powers $P_{n\sigma}$ and $P_{\sigma m}$ are emitted by the sample. We must emphasize that here P_{ii} are radiation powers absorbed or emitted by the paramagnetic sample, since we have used only the solutions of the matter equation. If we find the powers absorbed or emitted by the device, we should also take into account the field equations³ by considering the characteristics of the cavity. Thus, the quality factors corresponding to the emitted fields and p^2 in Eqs. (7) will be chosen so that the maser device will be at threshold for the emitted field of frequency ω_{am} (when a such emission is strong enough to balance all the losses of radiation), and above threshold for the emitted field of frequency $\omega_{n\sigma}$. In this case, the bridge device is balanced for the "idler" field of frequency $\omega_{\sigma m}$ and, at the same time, is a low-noise radiation source of frequency $\omega_{n\sigma}$ corresponding to a maser action with inversion, while p^2 defines the operation point of the device (see Fig. 1). The emitted field of frequency $\omega_{n\sigma}$ could be a reference field for a phase-sensitive detection of small signals of lower frequency $\omega_{n\sigma}$, while the radiation of frequency ω_{nm} will be the pump. Let it be a small signal of lower frequency $\omega'_{n\sigma} = \omega^o_{n\sigma} + d\omega'$, and let us suppose that this field and the reference field of frequency

 $\omega_{n\sigma} = \omega_{n\sigma}^o + d\omega$ corresponding to the emitted power $P_{n\sigma}$ are entirely coherent. That means the spectral density and the relative phase factor f is the same for every $\omega_{n\sigma} = \omega'_{n\sigma}$. In this case $d\langle p_{n\sigma}^2 \rangle$ in Eq. (4) is

$$d\langle p_{n\sigma}^2 \rangle = \langle p_{n\sigma}^2 \rangle - \langle p_{n\sigma}^{o2} \rangle \simeq 2f \langle p_{n\sigma}^o dp_{n\sigma} \rangle , \qquad (8)$$

where $p_{n\sigma}^{o}$ corresponds to the reference field, while $dp_{n\sigma}$ corresponds to the small detected signal. In this case, the bridge device becomes unbalanced and for a suitable sign of the phase factor, a power $dP_{\sigma m}$ corresponding to the observing field will be emitted by a maser action without population inversion.

Let dP_{ij} be the power changes as a consequence of the phase-sensitive detection of small signals of lower frequency $\omega_{n\sigma}$. Taking Eqs. (5) and (6) into account, $|dP_{n\sigma}| \ll |dP_{\sigma m}|, |dP_{nm}|$, so that $dP_{nm} \cong -dP_{\sigma m}$. Consequently, the emitted power corresponding to the observing field will be absorbed from the pump.

The amplification method described here seems to be similar to that used in a lower sideband up-converter paramp, where $\omega_{n\sigma}$ should be the signal frequency, $\omega_{\sigma m}$ the idler frequency, and ω_{nm} the pump frequency.^{4,6} However, there are essential differences between these methods. Thus, although the two methods entail a pump source, in the parametric amplification only one strong field (the pump) is involved; while in our case, three strong fields should be necessary. Two of them, the reference field and idler field, being emitted by maser actions with inversion and without inversion, respectively. In addition, this method corresponds to a phase-sensitive linear amplification. That is why the SB method, which combines the maser action and the parametric amplification, is rather similar to that used in a transistor (triode), where the level n is the emitter (the cathode), level σ is the common base (the control grid), and level m is the collector (the anode) (see Fig. 1).



FIG. 1. Power characteristics (in arbitrary units) of the "quantum transistor," where $\delta_2 = (\langle p_{nm}^{o2} \rangle - \langle p_{n\sigma}^{o2} \rangle) / \langle p_{n\sigma}^{o2} \rangle$, and where the operating point of the amplifier is $(p^2/\langle p_{n\sigma}^2 \rangle) \approx 0.04$. P_{nm} , $P_{\sigma m}$, and $P_{n\sigma}$ correspond to the pump, idler, and reference fields, while $dp_{n\sigma}$ and $dP_{\sigma m}$ correspond to the signal and observing fields.

The added noise by the quantum amplification is essential for the performance of linear amplifiers. As the three fields (pump, reference, and idler) are all strong, they are assumed to be excited in large amplitude coherent states, so that they can be regarded as classical;⁵ they remain unaffected by their coupling to the signal and to the observing field. That is, the noise added by the present method could be treated in a similar way to that described by Caves for linear amplifiers.¹

It is interesting to note the optimum conditions that must be fulfilled to obtain a maximum amplification. Thus, the choice of the dilute paramagnetic crystal is very important. $(T_2^{ij})^{-1}$ in our treatment represents the homogeneous broadening mechanisms of a simple line; consequently, the inhomogeneous broadening mechanisms have to be negligible. As a consequence of the dominant isotropic character of the electronparamagnetic-resonance spectra, the atoms and ions with ns' unpaired electrons, and molecule ions with σ unpaired electrons and large hyperfine interactions, stabilized in suitable host lattices with a low concentration of isotopes with nonzero nuclear spin, could be in our case, ideal spin systems. Unlike the double-resonance maser action where, in the linear case, the amplification is independent on the spin-lattice relaxation times T_1 and linear dependent on T_2 ,³ in the present case, the amplification is linear dependent on $(T_1)^{-1}$ and on $(T_2)^2$:

$$dP_{\sigma m} \sim (T_2 / T_1) T_2 N , \qquad (9)$$

where we have considered in Eq. (4) that T_2^{ij} are all comparable with T_2 [see Eq. (10)], while $T_{ij}^{jr} = a_{ij}^r T_1$ are all comparable with T_1 and have the same temperature dependence (because a_{ij}^r are temperature independent). In addition, for a multilevel spin system $T_1^{ij} > T_2^{ij}$.^{7,8} On the other hand,⁷⁻¹¹

$$(T_2^{ij})^{-1} = (T_2^s)^{-1} + b_{ij}(T_1)^{-1}, (10)$$

where for a multilevel spin system $b_{ij} \gtrsim 1$ are temperature independent, T_2^s represents the usual spin-spin relaxation mechanisms, and is temperature independent and linear dependent on the spin concentration C (Ref. 9),

$$(T_2^s)^{-1} = \alpha C$$
 (11)

Because b_{ij} are all comparable, we can consider in Eq. (10) for simplicity $b_{ij} \approx 1$. In this case [see Eq. (9)], since $N \sim C$: $dP_{\sigma m} \sim x (1+x)^{-2}$, where $x = (T_2^s/T_1)$. As the function $x (1+x)^{-2}$ exhibits a maximum for x = 1, the amplification has a maximum when the spin-spin and spin-lattice relaxation rates are comparable. For the spin systems mentioned above, the inhomogeneous broadening contribution to the linewidth is usually $(T_2^*)^{-1} \leq 0.1$ G. As $(T_2)^{-1}$ have to be much larger than $(T_2^*)^{-1} \leq 0.1$ G. As $(T_2)^{-1}$ have to be much larger than $(T_2^*)^{-1}$, and α in Eq. (11) is for S electron atoms and ions or molecular radicals of about $(3-8) \times 10^{-13}$ cm³/s,⁹ concentrations of about $(10^{18}-10^{19})$ cm⁻³ should be suitable. Consequently, for an optimum amplification, T_1 must have an optimum value, which is expected to be of about $(10^{-6}-10^{-7})$ s. In this case, the Raman spin-lattice relaxation mechanisms are dominant and strongly temperature dependent.^{10,11} That is why an optimum

amplification will be achieved at an optimum temperature, which for S-electron ions with large spin-orbit interactions in the avaited states (Ph^{3+} Hc^+ Tl^{2+}) (Pefe

ture, which for S-electron ions with large spin-orbit interactions in the excited states (Pb^{3+} , Hg^+ , Tl^{2+}) (Refs. 11–13) is expected to be about 80–100 K, while for H^0 (Ref. 9) or molecular ions with σ unpaired electrons,¹⁴ even more elevated than 100 K.

The spin-spin relaxation rate $(T_2^s)^{-1}$ (and consequently the optimum spin-lattice relaxation rates) increases with increasing spin concentration. That is, the higher spin concentrations, i.e., the higher optimum temperatures and pump power levels, and the larger bandwidths [see Eq. (2)].

Let us compare the power gain of this amplification with the usual maser action. Thus, in the case of doubleresonance maser action with inversion,³ the lower the temperature, the better the amplification. Consider, for example, that the pumping frequency is ω_{nm} and the emitted power $dP'_{n\sigma}$ corresponds to a small signal of frequency $\omega_{n\sigma}$ mentioned above. In this case, the relative efficiency of the amplification proposed here as compared to that of the usual maser action⁸ is

$$\varepsilon^{r} = dP_{\sigma m} / dP'_{n\sigma}$$

$$\approx (\omega_{\sigma m} / \omega_{n\sigma}) (T_{L} / T_{op}) (\langle p_{n\sigma}^{o} dp_{n\sigma} \rangle / \langle dp_{n\sigma}^{2} \rangle) \gg 1 , \qquad (12)$$

where T_L is the low temperature at which operates a usual maser, $T_{\rm op}$ is the optimum temperature corresponding to the maximum power gain of the SB amplification method, and where the ratios $(T_L/T_{\rm op})$ and $(\omega_{n\sigma}/\omega_{\sigma m})$ could be considered comparable.

The major differences between the SB amplification method and the usual maser action or parametric amplification are (a) unlike the two methods, which are both phase-insensitive and have comparable power gain, the SB method is phase sensitive, has a predicted power gain much larger, and is based on a maser action without inversion. As has been shown,¹⁵ the phase noise added by a phase-sensitive amplification can be reduced to such an extent that, in the limit of large amplification, it can be much less than the phase noise of a coherent state. (b) In contrast to the usual maser action for which the ambient temperature is very low, in our case the amplification exhibits a maximum at an optimum temperature which is relatively high. By a suitable choice of the spin concentration (which in the case of S electron atoms and ions or molecular radicals can be easily modified by irradiation or thermal annealing), this optimum temperature could be easily changed. Consequently, from this point, the SB method is similar to the parametric amplification. (c) At this optimum temperature, the spin-lattice relaxation times are very short—about $(10^{-6}-10^{-7})$ s. Thus, although we have used steady-state solutions, for changing rates of the detected signal amplitudes or of the phase factor up to $(10^5 - 10^6)$ s⁻¹, these solutions could remain valid (the quasistationary case). In addition, by a suitable phase switching, short low-noise pulses in the microwave or the far-infrared region could be obtained. (d) The bandwidth is not expected to be as narrow as in the maser case. The higher the spin concentration, the larger the bandwidth will be. (e) The pump power level is expected to be much higher than in the usual maser action, and even higher than that used in a paramp. However, the stronger the pump field, the better the amplification [see Eq. (12)]. (f) Unfortunately, as in the usual maser case, and unlike the paramp case, the pump frequency and the magnetic field is fixed by the operating frequencies and the crystal used. In addition, as in the paramp case and unlike the maser case, the gain sensitivity depends strongly on the pump-field intensity. However, by using in parallel a similar bridge, so that this bridge maintains balance with respect to the idler field, the pump-field intensity as well as the dc magnetic field could be well stabilized.

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