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PROPOSAL FOR AMPLIFICATION WITHOUT INVERSION CONSIDERING THE SIMPLE SCHEME OF DOUBLE RESONANCE

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It is shown that amplification without the population inversion may be possible even in the usual case of maser action, that corresponds to a strong pumping field and a weak signal, both at resonance. The optimum conditions for such amplification are established by means of the steady-state solutions of the density matrix for multilevel spin system in dilute paramagnetic crystals at high temperatures.

1. Introduction

Amplification without population inversion has recently attracted considerable attention. Many schemes for light amplification without population inversion have been proposed [1], while some of them have already been experimentally demonstrated [2].

Microwave amplification without the population inversion has also been recently predicted [3]. In the case of triple resonance, when the spectroscopic bridge conditions are satisfied, important linear and nonsaturating effects are predicted. Therefore, high-efficiency microwave generation or phase-sensitive detection could be achieved [3,4]. In the usual case of double resonance such effects may not be possible [3]. However, the amplification without the population inversion, in the usual case of a strong pumping field and a weak signal, both at resonance, would have the advantage of being a simple scheme, while all the other schemes proposed or achieved until now are more complicated.

The purpose of this paper is to establish the optimum conditions of amplifications without inversion corresponding to the usual scheme of maser action mentioned above.

2. Formalism

In order to describe the microwave powers absorbed or emitted by a dilute paramagnetic solid subjected to several quasimonochromatic fields at resonance, one can use the analytic steady-state solutions of the density matrix equation for multilevel spin system [3,4]:

$$P_{ij} = \int P_{ij}(\omega) D_{ij}(\omega) \mathrm{d}\omega = 2\hbar\omega_{ij}^0 N \Omega_{ij}. \tag{1}$$

If $P_{ij} > 0$, the power is emitted, while if $P_{ij} < 0$, the power is absorbed. $\omega_{ij}^0 = \omega_i^0 - \omega_j^0$, $\omega_i^0 = E_i/\hbar$, with E_i being the exact eigenvalues of the spin Hamiltonian and N the total number of spins. $\omega_{ij} = \omega_{ij}^0 + \delta\omega$, where the spectral density $D_{ij}(\omega)$ of the quasimonochromatic field at resonance is nonvanishing as long as $\mid \delta\omega \mid \ll (T_2^{ij})^{-1}$, where T_2^{ij} is the spin-spin relaxation time corresponding to the pair of levels involved.

Let us consider three levels n, σ, m among the levels of a multilevel spin system. Let us suppose that the number of levels is $N > 2$, $E_n > E_\sigma > E_m$ and that ω_{nm}^0, $\omega_{n\sigma}^0$, are the frequencies of two quasimonochromatic fields at resonance. ω_{nm} corresponds to a strong pumping field, while $\omega_{n\sigma}$ corresponds to a weak signal. In our case $P_{nm} < 0$ and $P_{n\sigma} > 0$ correspond, respectively, to the absorption and emission of microwave power by the lattice. The solutions for the power parameters Ω_{nm} and $\Omega_{n\sigma}$ ($\Omega_{ij} = -\Omega_{ji}$) in Eq. (1) are obtained by solving a system of equations [3,4], which in our case becomes

$$\{\left[(T_2^{nm})^{-1} + < p_{n\sigma}^2 > T_2^{\sigma m}\right] < p_{nm}^2 >^{-1} + 2T_1^{nm}\}\Omega_{mn} - (T_2^{\sigma m} + 2T_{n\sigma}^{mn})\,\Omega_{n\sigma} =$$
$$= \rho_{mm}^0 - \rho_{nn}^0 \tag{2a}$$

$$\{\left[(T_2^{n\sigma})^{-1} + < p_{nm}^2 > T_2^{\sigma m}\right] < p_{n\sigma}^2 >^{-1} + 2T_1^{n\sigma}\}\Omega_{n\sigma} - (T_2^{\sigma m} + 2T_{mn}^{n\sigma})\,\Omega_{mn} =$$
$$= \rho_{nn}^0 - \rho_{\sigma\sigma}^0 \tag{2b}$$

where ρ_{ii}^0 are the thermal equilibrium values of the diagonal elements of the density matrix and $\rho_{ii} = N_i/N$, N_i being the population of the spin level i. p_{ij}

are the matrix elements (considered to be real) of the time dependent Hamiltonian which represents the interactions of the multilevel spin system with the microwave fields. They are expressed in units of $\hbar$ and are written in the interaction representation like the off-diagonal elements ρ_{ij} of the density matrix. Further, $< p_{ij}^2 >= \int p_{ij}^2(\omega_{ij}) D_{ij}(\omega)\mathrm{d}\omega$. T_2^{ij} is the spin-spin relaxation time corresponding to the pair of spin levels i, j, while T_1^{ij} and $T_{ij}^{\alpha i}$ are spin-lattice relaxation times defined as:

$$T_1^{ij} = T_{ji}^{ij} = K_{ij}^j - K_{ij}^i, \tag{3a}$$

$$T_{ij}^{\alpha i} = K_{ij}^\alpha - K_{ij}^i \tag{3b}$$

where T_1^{ij} represents the spin-lattice relaxation time corresponding to a simple line in usual electron paramagnetic resonance (EPR) experiments [5,6]. The "spin-lattice relaxation times" K_{ij}^α are obtained by solving the equations:

$$\sum_{\alpha\neq i}\left(K_{ij}^\alpha w_{\alpha i} - K_{ij}^i w_{i\alpha}\right) = \sum_{\alpha\neq j}\left(K_{ij}^j w_{j\alpha} - K_{ij}^\alpha w_{\alpha j}\right) = 1;$$

$$\sum_{\alpha\neq\beta}\left(K_{ij}^\beta w_{\beta\alpha} - K_{ij}^\alpha w_{\alpha\beta}\right) = 0 \quad \beta\neq i, j \tag{4}$$

where $w_{i,j}$ are the spin-lattice relaxation rates corresponding to the pair of states i, j.

The solution for ρ_{ii} (corresponding to the population of the level i) becomes

$$\rho_{ii} = \rho_{ii}^0 + 2K_{nm}^i\Omega_{nm} + 2K_{n\sigma}^i\Omega_{n\sigma}. \tag{5}$$

The equations (1), (2) and (4) are valid as long as [3,6]:

$$\frac{< p_{ij}^2 >}{R_{\alpha\beta}R_{ij}} \ll 1 \tag{6}$$

where $-R_{\alpha\beta}$ are relaxation rates corresponding to the pairs of different vibrational states and representing the line-widths in the infrared (IR) spectra. As $-R_{ij} = (T_2^{ij})^{-1}$, which correspond to the EPR line-width, is of few G (10^{-4} cm^{-1}), the condition (6) is satisfied even for very strong microwave fields, whose magnetic amplitudes do not exceed a few hundreds G.

An improved amplification is achieved when the linear effects prevail [7] when the emitting power is proportional to the intensity of the emitted field. This intensity, in the case of double resonance [3,4], has to be much lower than the intensity of the saturating field. Therefore,

$$< p_{n\sigma}^2 > \ll < p_{nm}^2 > . \tag{7}$$

Let us suppose that the saturating field is strong enough so that:

$$T_2^{nm} T_2^{\sigma m} < p_{nm}^2 > \gg 1. \tag{8}$$

Taking into account the conditions (7) and (8) in Eqs. (1), (2) and (4) one obtains:

$$\rho_{nn} - \rho_{mm} \approx \left(\rho_{nn}^0 - \rho_{mm}^0\right) (T_2^{nm})^{-1} (T_1^{nm})^{-1} < p_{nm}^2 >^{-1} \approx 0 \tag{9}$$

$$\rho_{nn} - \rho_{\sigma\sigma} \approx \rho_{nn}^0 - \rho_{\sigma\sigma}^0 + (T_1^{nm})^{-1} T_{nm}^{\sigma n} (\rho_{mm}^0 - \rho_{nn}^0) \tag{10}$$

$$P_{n\sigma} \approx 2N\hbar\omega_{n\sigma} < p_{n\sigma}^2 >< p_{nm}^2 >^{-1} \left[(\rho_{mm}^0 - \rho_{nn}^0)(2T_1^{nm})^{-1} + (\rho_{nn}^0 - \rho_{\sigma\sigma}^0)(T_2^{\sigma m})^{-1} \right]. \tag{11}$$

In order to achieve the amplification without inversion, $\rho_{nn} - \rho_{\sigma\sigma}$ must be negative and $P_{n\sigma}$ positive. Therefore, the condition of amplification without inversion is:

$$\frac{T_{nm}^{\sigma n}}{T_1^{nm}} < \frac{\rho_{\sigma\sigma}^0 - \rho_{nn}^0}{\rho_{mm}^0 - \rho_{nn}^0} < \frac{T_{nm}^{\sigma n}}{T_1^{nm}} + \frac{T_2^{\sigma m}}{2T_1^{nm}}. \tag{12}$$

3. *Discussion*

At low temperatures, the spin-spin relaxation times are usually much shorter than the spin-lattice relaxation times, i.e., $T_2^{\sigma m} \ll T_1^{nm}$. In that case the condition (12) would not be fulfilled and the amplification without inversion could not be achieved.

At higher temperatures, the spin-spin and spin-lattice relaxation times become comparable and the EPR line-widths in some cases have strong temperature dependence [8]. The larger the ratio $T_2^{\sigma m}/T_1^{nm}$, the stronger is the amplification without inversion.

The spin-lattice relaxation times $T_{nm}^{\sigma n}$ and T_1^{nm} are always positive. As $T_1^{nm} = T_{nm}^{\sigma n} + T_{nm}^{m\sigma}$ (see Eq(3)) and $T_{nm}^{\sigma n}$ and $T_{nm}^{m\sigma}$ are both positive, the ratio $T_{nm}^{\sigma n}/T_1^{nm} < 1$. In the case of the multilevel spin systems, even T_2^{ij} and T_1^{ij} are comparable, $T_2^{ij} < T_1^{ij}$, so that (see Eq. (12)),

$$\frac{\rho_{\sigma\sigma}^0 - \rho_{nn}^0}{\rho_{mm}^0 - \rho_{nn}^0} \approx \frac{E_n - E_\sigma}{E_n - E_m} \lesssim 1.$$

Therefore, the frequency of the pumping field have to be higher than that of the emitted field. At temperatures that are not too high $(\rho_{ii}^0 - \rho_{jj}^0) \sim (1/T)$, T_2 is usually temperature independent, while $(T_1)^{-1} \sim T^7$ or increases exponentially with temperature [8,9]. At sufficiently high temperatures $(T_2)^{-1}$ and $(T_1)^{-1}$ are both proportional to the temperature [8]. Since $\rho_{nn} - \rho_{\sigma\sigma}$ would have to be negative, the larger the value of $\rho_{\sigma\sigma} - \rho_{nn}$, the weaker is emitted field $P_{n\sigma}$ (see Eq. (11)). That is why $P_{n\sigma}$ and the amplification without inversion exhibits a maximum at an optimum temperature. On the contrary, in the usual case of amplification (with inversion), the lower the temperature, the better is the amplification [3,7].

Consequently, in order to achieve the amplification without the population inversion, sufficiently high temperatures are needed, so that the spin-spin and spin-lattice relaxation rates become comparable. In other words, at relatively low temperatures, when the spin-spin relaxation times are much shorter than the spin-lattice relaxation times, the spins relax to equilibrium inside the spin system, and after that, the spin reservoir relaxes to the lattice. In that case the amplification without the population inversion is not possible. At sufficiently high temperatures, when the spin-spin and spin-lattice relaxation times become comparable, the spins relax among themselves and to the lattice with comparable rates. In this case, when the conditions (6), (7), (8) and (12) are fulfilled, the amplification without the population inversion becomes possible, even in the usual case of double resonance, but only when the two transition have a level in common [3].

The above considerations could be valid for any kind of multilevel system, including the usual case when the electric field contributes to p_{ij} [10], but only when all the conditions mentioned above are satisfied. Thus, the condition (6) and consequently the validity of Eqs. (1), (2) and (4) is specifical only for a spin system corresponding to a well isolated orbitally nondegenerate ground state [5,11]. That is why, for the spin system mentioned above, the conditions (6) and (8) may be satisfied simultaneously, and consequently our treatment may be valid, although the pumping field is very strong (see the condition (8)). For the amplification with inversion, the saturating condition for the pumping field is [5]: $T_1^{nm} T_2^{nm} < p_{nm}^2 > \gg 1$. At low temperatures $T_1^{nm} \gg T_2^{nm}$ and the condition (8) ceases to be necessary. That is why our treatment might be inadequate in the case of multilevel systems other than spin systems in dilute paramagnetic solids.

Finally, we have to emphasize that, although the scheme analysed in this paper does not exhibit some advantages over those corresponding to the spectroscopic bridge (the triple resonance case [3]), it is the simplest scheme of amplification without inversion predicted or realized until now.

References

1) For reviews see: O. Kocharosckaya, Phys. Rep. **219** (1992) 175; M. O. Scully, Phys. Rep. **219** (1992) 191;

2) J. Gao, C. Guo, X. Guo, G. Jin, P. Wang, J. Zhao, H. Zhang, Y. Jiang, D. Wang and D. Jiang, Opt. Commun. **93** (1992) 323; A. Nottelmann, C. Peters, and W. Lange, Phys. Rev. Lett. **70** (1993) 1783; W. E. van der Veer, R. J. J. van Diest, A Donszelmann and H. B. van Linden van der Heuvell, Phys. Rev. Lett. **70** (1993) 3243;

3) F. F. Popescu, Phys. Rev. B **48** (1993) 13569;

4) F. F. Popescu and F. Marica, Romanian Rep. Phys. (to be published);

5) F. F. Popescu (to be published);
6) A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, 1961), pp. 526;
7) W. Orton, D. H. Paxman and J. C. Walling, *The Solid State Masers* (Pergamon, Glasgow, 1970);
8) F. F. Popescu and V. V. Grecu, Phys. Stat. Solidi b **68** (1975) 595, and J. Phys. C **15** (1982) 1547;
9) A. Abragam and B. Bleany, *Electron Paramagnetic Resonance of Transition Ions* (Claredon, Oxford, 1970);
10) C. Feuillade, J. C. Baker and C Bottcher, Chem. Phys. Lett. **40** (1976) 121;
11) F. F. Popescu and V. V. Grecu, J. Phys. C **15** (1982) 1531.

PRIJEDLOG POJAČANJA BEZ INVERZIJE POMOĆU JEDNOSTAVNE SHEME DVOSTRUKE REZONANCIJE

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Pokazano je da je pojačanje bez populacijske inverzije moguće čak i u uobičajenom slučaju maserske akcije koja odgovara jakom pumpajućem polju i slabom signalu, oboje u rezonanciji. Optimalni uvjeti za takvo pojačanje nađeni su kao stacionarno rješenje matrice gustoće za spinski sistem s više nivoa u razrijeđenim paramagnetskim kristalima na visokim temperaturama.