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NONLINEAR PROPERTIES OF THREE-LEVEL SYSTEMS

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As is well known, the three-level system* is the basic element of quantum amplifiers and oscillators-masers. It is of interest to consider other possible physical applications of three-level systems, in particular, applications that derive from their nonlinear properties.

A manifestation of the nonlinear properties of a three-level system would be the response of the system (for example, the polarization P) to two monochromatic signals. Let E_1 , E_2 and E_3 be the three levels of the quantum system and suppose that an external field (electric or magnetic) acts on the system

$$F = E_{13} \cos \Omega_{31} t + E_{23} \cos \Omega_{32} t, \quad (1)$$

where $\Omega_{31} \approx (E_3 - E_1)/\hbar$ and $\Omega_{32} \approx (E_3 - E_2)/\hbar$.

To find the system polarization produced by the field (1) we use the equation for the density matrix ρ_{mn} :^[1,2]

$$\begin{aligned} \frac{\partial \rho_{mn}}{\partial t} + i\omega_{mn}\rho_{mn} &= \frac{i}{\hbar} F \sum_{l=1}^3 (\mu_{ml}\rho_{ln} - \rho_{ml}\mu_{ln}) - [\tau^{-1}(\rho - \rho_0)]_{mn}; \\ [\tau^{-1}(\rho - \rho_0)]_{mn} &= \begin{cases} \tau_1^{-1}(\rho - \rho_0)_{mm} & \text{for } m = n \\ \tau_2^{-1}\rho_{mn} & \text{for } m \neq n \end{cases}, \end{aligned} \quad (2)$$

where the μ_{ml} are the dipole moment matrix elements, τ_1 and τ_2 are the longitudinal and trans-

verse relaxation times, and ρ_{0mn} is the density matrix corresponding to instantaneous equilibrium at time t , when the field is given by $F(t)$. The polarization of the system is

$$P = \text{Sp}(\hat{\rho}\hat{\mu}). \quad (3)$$

In solving Eq. (2) we keep only the resonance terms^[2] at frequencies Ω_{32} , Ω_{31} , and $\Omega_{31} - \Omega_{32}$, thereby obtaining the corresponding system of algebraic equations^[2] that yields the following expression:

$$P = \rho_{31}^- \mu_{13} e^{-i\Omega_{31}t} + \rho_{32}^- \mu_{23} e^{-i\Omega_{32}t} + \rho_{21}^- \mu_{12} e^{-i(\Omega_{31} - \Omega_{32})t} + \text{c.c.}, \quad (4)$$

if $\Omega_{31} = (E_3 - E_1)/\hbar$ and $\Omega_{32} = (E_3 - E_2)/\hbar$,

$$\rho_{31}^- = 2i\gamma_{31}\Delta^{-1} \{D_{13}^{(0)} [4(\tau_2^{-1} + \gamma_{23}^2\tau_1) + \tau_2\gamma_{13}^2]$$

$$- D_{23}^{(0)} (2\tau_1 + \tau_2)\gamma_{23}^2\},$$

$$\rho_{32}^- = 2i\gamma_{32}\Delta^{-1} \{D_{23}^{(0)} [4(\tau_2^{-1} + \gamma_{13}^2\tau_1) + \tau_2\gamma_{23}^2]$$

$$- D_{13}^{(0)} (2\tau_1 + \tau_2)\gamma_{13}^2\},$$

$$\rho_{21}^- = \frac{1}{2} i\tau_2\gamma_{13}\gamma_{23}(\rho_{32}^-/\gamma_{32} + \rho_{31}^-/\gamma_{13})$$

$$= -2\gamma_{13}\gamma_{23}\tau_2\Delta^{-1} \{D_{13}^{(0)} [2(\tau_2^{-1} + \tau_1\gamma_{23}^2) - \tau_1\gamma_{13}^2]$$

$$+ D_{23}^{(0)} [2(\tau_2^{-1} + \gamma_{13}^2\tau_1) - \tau_1\gamma_{23}^2]\};$$

$$\Delta = [4(\tau_2^{-1} + \gamma_{23}^2\tau_1) + \tau_2\gamma_{13}^2] [4(\tau_2^{-1} + \gamma_{13}^2\tau_1)$$

$$+ \tau_2\gamma_{23}^2] - (2\tau_1 + \tau_2)^2\gamma_{13}^2\gamma_{23}^2;$$

$$\gamma_{13} = \mu_{13}E_{13}/\hbar = \gamma_{31}, \quad \gamma_{23} = \mu_{23}E_{23}/\hbar = \gamma_{32};$$

where $D_{13}^{(0)}$ and $D_{23}^{(0)}$ are the corresponding equilibrium population differences in the levels.

It is evident from Eq. (4) that the response of the system to two monochromatic signals contains a term at the combination frequency $\Omega_{12} = \Omega_{13} - \Omega_{23}$; this term results from the nonlinearity of the system.[†] In particular, in the case of practical interest, where one of the applied signals is large (for example, $\gamma_{13}\tau_2 > 1$), it turns out that $|\rho_{12}|/|\rho_{23}| > 1$. This result indicates the possibility of building a quantum frequency converter with appreciable conversion gain. From the point of view of noise characteristics it would appear that the quantum converter can compete with quantum amplifiers. Special interest attaches to the case in which Ω_{12} is a very low frequency and can be used directly as the intermediate frequency.

We note that in principle it is possible for oscillations to occur at the frequencies Ω_{13} and Ω_{23} when a three-level maser is used as an oscillator or as an amplifier. It then follows that a maser can produce a signal at frequency Ω_{12} even if the populations in levels E_1 and E_2 are not inverted.

The principle of quantum frequency conversion can also be used in the optical region. It would appear that the monochromatic light signals re-

quired for this purpose could be generated by lasers. In this connection we note that lasers often generate several frequencies rather than a single frequency.^[7,8] One expects that combination frequencies would be generated under these conditions.

*The idealization in which we consider only three levels of the entire system of levels is completely justified since the other levels can be neglected in most cases.

†The Bloembergen equations,^[3] in which the off-diagonal elements of the density matrix are not considered, are frequently used in maser calculations. It will be evident that these equations can not give the effect considered here nor a number of other features of maser operation. The reference made by Bloembergen^[4] to the calculation by Clogston,^[5] in which off-diagonal terms in the density matrix are included, is not well taken because the latter work^[5] contains an error in that the author has not taken account of all the resonance terms in the density matrix. A similar error appears in a paper by one of the present authors.^[6]

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NEGATIVE CONDUCTIVITY IN INDUCED TRANSITIONS

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A number of methods for producing negative temperatures in semiconductors^[1-3] have been proposed in recent years.

It has been noted^[3] that in indirect transitions the concentration of carriers for which a negative temperature is produced with respect to interband transitions is relatively small, being several orders of magnitude smaller than the concentration at which a negative absorption coefficient is obtained for photons with energies comparable to the width of the forbidden band. In order to obtain a negative absorption coefficient the probability for induced emission of photons in an interband transition must be appreciably greater than the probability of photon absorption in the inverse process, so as to compensate for absorption in internal transitions. However, internal absorption processes have essentially no effect on conductivity since they cause no change in the total number of free carriers; on the other hand, in-

duced interband transitions due to photons incident on a semiconductor in a negative temperature state reduce the number of free current carriers and cause a reduction in conductivity.

Thus, a semiconductor in a negative temperature state with respect to an interband transition should exhibit a negative photoconductivity when irradiated by photons with energies approximately equal to the width of the forbidden band. Measurement of the spectral dependence of the photoconductivity of a semiconductor should allow us to find negative temperature states even if a negative absorption coefficient is not produced.

We have carried out experiments to produce and observe negative temperatures in silicon. Samples maintained at a temperature of 4° K were irradiated by light at wavelengths smaller than 0.7 μ and exhibited an appreciable increase in conductivity. When irradiated by weak monochromatic light in a narrow wavelength band close to 1.1 μ, however, a number of samples exhibited reduced conductivity (negative photoconductivity).

It can be assumed that this reduction in conductivity is due to a negative temperature state, although we have not completely ruled out other explanations for the observed effect, for example, impurity photoconductivity.

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