

## Theory of Optical Beating in Photoconductors (Phenomenological Modification of Semiclassical Treatment)\*

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Recent laser experiments have shown that two coherent light waves of slightly different frequency, when simultaneously incident on a photodetector, produce a photosignal modulated at the beat frequency  $\Delta f$ . The theory of this photomixing process is developed by means of a modification of the usual semiclassical treatment of radiation. This modification takes into account the finite lifetime  $\tau$  of excited electronic states in the quantum-mechanical equations of motion. The photosignal is found to be proportional to  $[1 + 4\pi^2\tau^2(\Delta f)^2]^{-1/2}$ . This result suggests an experimental means of determining the lifetime by measuring the frequency response of the photosignal to the beat frequency. The theory compares favorably with experiments on *p*-type extrinsic gold-doped germanium.

### I. INTRODUCTION

RECENT experiments<sup>1</sup> with lasers have shown that two coherent light beams of slightly different frequency, when simultaneously incident upon a photodetector, produce a photosignal which is modulated at the difference frequency.

We examine the theory of this photomixing process in a detector in which the photosignal is proportional to the number of photoexcited electrons (e.g., photoconductor operated in Ohmic range). We review first the so-called semiclassical treatment of radiation and its application to the absorption of incoherent light by matter. Then, we show the complications which arise in an attempt to apply the same theory to coherent light.

These complications are avoided by modifying the usual semiclassical treatment in a phenomenological way, so as to take account of the finite lifetime of excited electronic states directly in the quantum-mechanical equations of motion.<sup>2</sup>

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<sup>1</sup> See, e.g., G. Lucovsky, M. E. Lasser, and R. B. Emmons, *Proc. IEEE* **51**, 166 (1963); M. DiDomenico, Jr., R. H. Pantell, O. Svelto, and J. N. Weaver, *Appl. Phys. Letters* **1**, 77 (1962); A. Javan, W. R. Bennet, and D. R. Herriott, *Phys. Rev. Letters* **6**, 106 (1961); C. M. Penchina and H. Levinstein, *Bull. Am. Phys. Soc.* **9**, 237 (1964).

<sup>2</sup> We proposed this modification in a paper presented at the St. Louis Meeting of the American Physical Society, 25–28 March 1963 [C. M. Penchina, *Bull. Am. Phys. Soc.* **8**, 234 (1963)]. As a result of that paper, we received communications from the respective authors indicating their treatments of photomixing which have since been published. They are: P. S. Pershan and N. Bloembergen, *Appl. Phys. Letters* **2**, 117 (1963); G. J. Lasher and A. H. Nethercot, Jr., *J. Appl. Phys.* **34**, 2122 (1963). Pershan and Bloembergen use quantum mechanics in the density matrix formulation to study the excitation of photoelectrons in a metal but do not treat the recombination process. Lasher and Nethercot, Jr., use quantum mechanics in the density matrix formulation to calculate the excitation of electrons in an intrinsic bulk photoconductor, but treat the recombination process phenomenologically using a rate equation with an assumed lifetime.

Our present treatment uses ordinary perturbation theory in a one-electron approximation and is simply a phenomenological modification of the usual semiclassical treatment of radiation.

Our result differs from that of Lasher and Nethercot, Jr., who find a dependence of the number of excited electrons on the intraband scattering time. As one is interested in the total number

This study of absorption of coherent light indicates a new technique for determining the lifetimes of excited states in photoconductors by measuring the frequency response of the photosignal to the beat frequency between two coherent light beams. This experimental technique is discussed by Penchina and Levinstein.<sup>3</sup>

### II. PHOTOCONDUCTIVITY

In an Ohmic photoconductor with a steady bias voltage applied, the photosignal is proportional to the number of optically excited electrons. In addition to the external radiation which excites electrons from their normal states, there are competing mechanisms which return the electrons to their normal states. These competing processes (e.g., radiative recombination, etc.) may, in general, be quite complicated to treat theoretically. However, in many cases of interest they are such that if the external radiation is turned off, the number of electrons in excited states decays exponentially to the equilibrium number.<sup>4</sup>

of excited electrons rather than their exact states within the band, and as intraband scattering changes, only the state within the band and not the total number of excited electrons, intraband scattering does not affect the result. As a rule the intraband scattering time is usually much shorter than the recombination time, thus the difference between the two results is usually quite difficult to measure experimentally. An interesting exception is the case of InSb at low temperatures. There, the intraband scattering time can be even longer than the recombination lifetime, and the difference between our two results would then be much more evident experimentally.

In a treatment inspired by that of Lasher and Nethercot, Jr., [C. M. Penchina, in *Proceedings of the 1964 International Conference on the Physics of Semiconductors* (to be published)] we use a completely quantum-mechanical approach in the density matrix formulation to study both the excitation and the recombination. This treatment allows for an actual calculation (at least formally) of the lifetime and leads to the same result as we get in our present phenomenological approach.

<sup>3</sup> Penchina and Levinstein, Ref. 1.

<sup>4</sup> In particular, this exponential decay does not occur, if the recombination is through traps. In such cases, the decay might sometimes be characterized by a sum of exponentials. We treat only those cases in which the decay is essentially exponential with one time constant. The effects of trapping in photoconductivity are widely discussed in the literature. For surveys and references, see, e.g., R. H. Bube, *Photoconductivity of Solids* (John Wiley & Sons, Inc., New York, 1960); R. A. Smith, *Semiconductors* (Cambridge University Press, Cambridge, England, 1959).

The usual means of treating the problem of photoconductivity makes use of a rate equation<sup>5</sup>

$$d(\delta n)/dt = G - R, \quad (\text{II.1})$$

in which one assumes the deviation from equilibrium is small so that the recombination rate  $R$  is simply  $\delta n/\tau$ . The generation rate is claimed to be proportional to the intensity of the incident light.<sup>6</sup> This claim is justified for incoherent light by means of the so-called semiclassical treatment of radiation,<sup>7</sup> which treatment ignores recombination. Thus the generation and recombination are treated independently, their combined effects being handled by means of the rate equation.

In Sec. III we apply this semiclassical treatment to coherent as well as incoherent incident radiation. It is seen that for coherent light, the justification of the assumed generation rate is no longer clear.

In order to get around this difficulty, we propose a modified semiclassical treatment of radiation, in which the recombination is no longer ignored in calculating the optical excitation. This modification allows for a direct calculation of the number of excess excited electrons as a function of time without using the rate equation as an intermediate step.

### III. SEMICLASSICAL TREATMENT OF RADIATION<sup>8</sup>

We treat the solid in a one-electron approximation.<sup>9</sup> We write the wave function as a linear superposition of a complete set of orthonormal eigenstates  $u_m$  of the field-free (unperturbed) Hamiltonian  $H_0$

$$\psi(t) = \sum_m a_m(t) u_m \exp(-i\omega_m t), \quad (\text{III.1})$$

where

$$H_0 u_m = E_m u_m = \hbar\omega_m u_m. \quad (\text{III.2})$$

In the presence of a radiation field, with interaction Hamiltonian  $\mathcal{H}$ , the Schrödinger equation yields

$$\dot{a}_j = (-i/\hbar) \sum_m a_m e^{i\omega_{jm}t} \mathcal{H}_{jm}, \quad (\text{III.3})$$

where

$$\mathcal{H}_{jm} = \langle u_j | \mathcal{H} | u_m \rangle$$

and

$$\omega_{jm} = \omega_j - \omega_m = (1/\hbar)(E_j - E_m).$$

To first order in the field strength

$$\mathcal{H} = (ie\hbar/mc) \mathbf{A} \cdot \nabla, \quad (\text{III.4})$$

<sup>5</sup> See, e.g., R. H. Bube, Ref. 4, pp. 63-64.

<sup>6</sup> Bube, Ref. 4, p. 64, footnote.

<sup>7</sup> This treatment is discussed in most textbooks on quantum mechanics; see, e.g., F. Seitz, *The Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), p. 215 ff; L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), p. 246 ff; D. Bohm, *Quantum Theory* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1951), p. 417 ff.

<sup>8</sup> See Ref. 7. Although the semiclassical treatment is widely discussed in many textbooks, we review it in this section to provide a basis for the section on applications. The application to coherent radiation is not treated in the standard texts on quantum mechanics.

<sup>9</sup> F. Seitz, Ref. 7, p. 234 ff, also p. 326.

and the probability that an electron which was initially in state  $u_I$  will be in state  $u_j$  at time  $t$  is, to first order,<sup>10</sup>

$$P_{jI}(t) = |a_{jI}(t)|^2 = \left| \frac{-i}{\hbar} \int_0^t e^{i\omega_{jI}t'} \mathcal{H}_{jI}(t') dt' \right|^2, \quad j \neq I, \quad (\text{III.5})$$

the additional subscript  $I$  indicating that the electron was initially in state  $u_I$ .

Let us note that the above approximation is valid only for times short enough that all  $a_m(t)$  change very little from their initial values. This may be a long time only if the perturbation is weak. Also, the time must be short compared with the lifetimes of the excited states in order that the results approximate the actual situation.

The probability that the electron is in any excited state is found by summing the above expression over all allowed (unfilled) excited states

$$P_I(t) = \sum_F P_{FI}(t). \quad (\text{III.6})$$

However, due to the time limitations on the validity of our approximation, one can not really hope to get a steady-state solution by this means. On the other hand, one can hope to find the rate of excitation per unit time at any time for which the  $a_m(t)$  have not changed much from the  $a_m(0)$ . This generation rate is then

$$\frac{dP_I(t)}{dt} = \sum_F \frac{dP_{FI}(t)}{dt}. \quad (\text{III.7})$$

The total rate of generation of excited electrons in the solid is found by summing over all the filled initial states  $u_I$ ,

$$\frac{dP(t)}{dt} = \sum_I \frac{dP_I(t)}{dt} = \sum_I \sum_F \frac{dP_{FI}(t)}{dt}. \quad (\text{III.8})$$

#### A. Application of Semiclassical Treatment

If the incident radiation propagates in the  $x$  direction and is polarized in the  $z$  direction

$$\mathbf{A} = \sum_n \mathbf{A}_n \exp i(\mathbf{k}_n \cdot \mathbf{r} - \omega_n t) + \text{c.c.}, \quad (\text{III.9})$$

where

$$\mathbf{A}_n = (0, 0, A_n), \quad A_n = |A_n| \exp(i\alpha_n),$$

$$\mathbf{k}_n = (k_n, 0, 0), \quad \omega_n = k_n c.$$

Thus

$$a_{FI}(t) = \frac{-i}{\hbar} \sum_n \sum_{m=1}^2 \mathcal{H}_{FI}^{nm} \times \frac{\exp\{i[\omega_{FI} + (-)^m \omega_n]t\} - 1}{\omega_{FI} + (-)^m \omega_n}, \quad (\text{III.10})$$

<sup>10</sup> We assume that  $u_j$  is a state in an unfilled band.

where

$$\begin{aligned}\mathcal{H}_{FI}^{n1} &= (ie\hbar/mc)A_n(\beta_n)_{FI}, \\ \mathcal{H}_{FI}^{n2} &= (ie\hbar/mc)A_n^*(\beta_n^*)_{FI}, \\ (\beta_n)_{FI} &= (u_F | e^{ik_n x} \partial / \partial z | u_I), \\ (\beta_n^*)_{FI} &= (u_F | e^{-ik_n x} \partial / \partial z | u_I).\end{aligned}$$

Taking the absolute square of the transition amplitude  $a_{FI}$ , to find the transition probability  $P_{FI}$ , leads to a large number of terms. These terms fall into two groups: direct terms, i.e., those which are the absolute squares of the individual terms in Eq. (III.10); and interference terms, i.e., those which are cross terms from taking the absolute square of the complete expression in Eq. (III.10). We find

$$P_{FI}(t)_{\text{direct}} = \frac{2}{\hbar^2} \sum_n \sum_{m=1}^2 |\mathcal{H}_{FI}^{nm}|^2 \times \frac{1 - \cos[\omega_{FI} + (-)^m \omega_n]t}{[\omega_{FI} + (-)^m \omega_n]^2} \quad (\text{III.11})$$

and<sup>11</sup>

$$P_{FI}(t)_{\text{interference}} = \frac{1}{\hbar^2} \sum'_{nm,ab} \mathcal{H}_{FI}^{nm} (\mathcal{H}_{FI}^{ab})^* W_{FI}^{nm,ab}, \quad (\text{III.12})$$

where

$$W \equiv \frac{1 - e^{i(\Omega+\theta)t} - e^{-i(\Omega-\theta)t} + e^{i2\theta t}}{(\Omega+\theta)(\Omega-\theta)},$$

$$\Omega_{FI}^{nm,ab} \equiv \omega_{FI} + \{ [(-)^m \omega_n + (-)^b \omega_a] / 2 \},$$

$$\theta^{nm,ab} \equiv [(-)^m \omega_n + (-)^b \omega_a] / 2.$$

The total transition probability is just the sum of these two sets of terms

$$P_{FI}(t) = P_{FI}(t)_{\text{direct}} + P_{FI}(t)_{\text{interference}}. \quad (\text{III.13})$$

### 1. Incoherent Radiation

If the sum of plane waves in Eq. (III.9) is an incoherent sum, i.e., if the  $\alpha_n$  are independent and random, the interference terms essentially cancel and

$$P_{FI}(t)_{\text{incoherent}} = P_{FI}(t)_{\text{direct}}. \quad (\text{III.14})$$

This is the case usually treated in textbooks. The well-known solution is

$$\frac{dP}{dt} = \sum_{n,F} \frac{2\pi e^2}{m^2 c^2} |(\beta_n)_{FI}|^2 |A_n|^2 \rho(\omega_I) \Big|_{\omega_{FI}=\omega_n}. \quad (\text{III.15})$$

If the radiation covers only a small range of frequencies

$$\sum'_{nmab} \quad \sum_n \sum_{m=1}^2 \sum_{a=b=1}^2$$

but exclude terms for which the ordered pair  $(nm)$  is equal to the ordered pair  $(ab)$  as they are the direct terms of Eq. (III.11).

so that for all  $n$

$$(\beta_n)_{FI} \approx (\beta_N)_{FI}, \quad k_n \approx k_N, \quad (\text{III.16})$$

then

$$dP/dt = DI, \quad (\text{III.17})$$

where

$$D = \frac{4\pi^2 e^2}{m^2 c \omega_N^2} \sum_F |(\beta_N)_{FI}|^2 \rho(\omega_I) \Big|_{\omega_{FI}=\omega_N}, \quad (\text{III.18})$$

where  $\rho(\omega_I)d\omega_I$  is the number of initially filled states in the energy range between  $\omega_I$  and  $\omega_I + d\omega_I$ , and where  $I$  is the classical intensity for the incoherent radiation.

In particular, if the allowed final states are  $N_A$  acceptor states, all with energy very close to  $E_A = \hbar\omega_A$ , we find

$$D = \frac{4\pi^2 e^2}{m^2 c \omega_N^2} N_A |(\beta_N)_{FI}|^2 \rho(\omega_I) \Big|_{\omega_F=\omega_A; \omega_I=\omega_A-\omega_N}. \quad (\text{III.19})$$

### 2. Coherent Radiation

If the sum of plane waves in Eq. (III.9) is a coherent sum, i.e., if the various phases  $\alpha_n$  have definite relations to one another (rather than being random), then we must consider the interference terms as well as the direct terms.

We find from Eq. (III.12)

$$\frac{dP_{FI} \text{ interference}}{dt} = \frac{1}{\hbar^2} \sum'_{nm,ab} \mathcal{H}_{FI}^{nm} (\mathcal{H}_{FI}^{ab})^* \frac{dW_{FI}^{nm,ab}}{dt}, \quad (\text{III.20})$$

where

$$\begin{aligned}\frac{dW}{dt} &= \frac{-i(\Omega+\theta)e^{i(\Omega+\theta)t} + i(\Omega-\theta)e^{i(\Omega-\theta)t} + 2i\theta e^{i2\theta t}}{(\Omega+\theta)(\Omega-\theta)} \\ &= e^{i2\theta t} \left\{ \frac{\sin(\Omega-\theta)t}{\Omega-\theta} + \frac{\sin(\Omega+\theta)t}{\Omega+\theta} \right. \\ &\quad \left. + \frac{i[1-\cos(\Omega-\theta)t]}{\Omega-\theta} - \frac{i[1-\cos(\Omega+\theta)t]}{\Omega+\theta} \right\}. \quad (\text{III.21})\end{aligned}$$

Let us examine  $dW/dt$  more closely. The two terms with sines in the numerator are functions just like those which arise for incoherent light<sup>7</sup> and are considered as Dirac delta functions with respect to any slowly varying functions. The other two terms are

$$g(\Omega, \theta, t) = i \left[ \frac{1 - \cos(\Omega - \theta)t}{\Omega - \theta} - \frac{1 - \cos(\Omega + \theta)t}{\Omega + \theta} \right]. \quad (\text{III.22})$$

This expression is finite everywhere. At the zeros of the denominator it becomes

$$g(\pm\theta, \theta, t) = -i[1 - \cos 2\theta t] / 2\theta; \quad (\text{III.23})$$

and for  $\Omega \neq \pm\theta$  and  $t$  large,  $g(\Omega, \theta, t)$  oscillates rapidly as a function of  $\Omega$ . One might thus be tempted to say that,

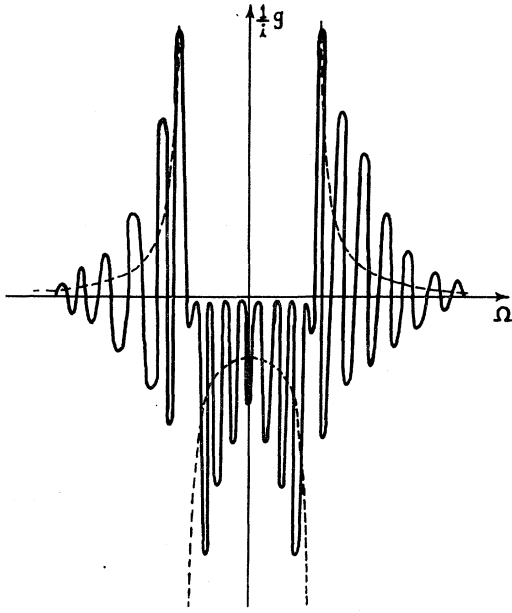


FIG. 1. Schematic sketch of  $g(\Omega, \theta, t)$ . Dotted curve shows time average.

in general, for sufficiently large  $t$  and arbitrary  $f(\Omega)$

$$\int_{-\infty}^{\infty} f(\Omega) g(\Omega, \theta, t) d\Omega \xrightarrow[t \rightarrow \infty]{?} 0. \quad (\text{III.24})$$

However, combining terms in Eq. (III.22), we find

$$g(\Omega, \theta, t) = \frac{2i[\theta - \theta \cos \Omega t \cos \theta t - \Omega \sin \Omega t \sin \theta t]}{\Omega^2 - \theta^2}, \quad (\text{III.25})$$

which, we see, oscillates about an average value of  $2i\theta/(\Omega^2 - \theta^2)$  rather than oscillating about the  $\Omega$  axis (see schematic sketch in Fig. 1). It is then obvious by examination that Eq. (III.24) is not valid in general. [For an example of a simple function which violates the conjecture of Eq. (III.24), see Appendix A.]

It is interesting to note, however, that if we neglect  $g(\Omega, \theta, t)$  in Eqs. (III.20) and (III.21) (and that is quite a big if), then we find, using approximations similar to those used in the preceding treatment of incoherent light, that

$$dP(t)/dt = DI(t), \quad (\text{III.26})$$

where  $I(t)$  is the classical intensity as a function of time, and  $D$  is given by Eq. (III.18) or (III.19). The actual calculations are performed in Appendix B.

We emphasize that the result of Eq. (III.26) is based on the false assumption that the conjecture of Eq. (III.24) is valid. If, on the other hand, we do not neglect  $g(\Omega, \theta, t)$ , then we are unable to draw any simple qualitative general conclusions, but require very detailed information about the particular system of interest.

Now, in order to take account of the finite lifetime  $\tau$  of the excited electronic states of the photoconductor,

we substitute Eq. (III.26) into the rate equation, Eq. (II.1). We find in the steady state

$$\begin{aligned} \delta n = & \frac{Dc\tau}{2\pi} \sum_{n,a} k_n k_a |A_n| |A_a| \\ & \times \left\{ \frac{\cos[(\omega_n - \omega_a)t - (\alpha_n - \alpha_a) - \tan^{-1}(\omega_n - \omega_a)\tau]}{[1 + (\omega_n - \omega_a)^2 \tau^2]^{1/2}} \right. \\ & \left. - \frac{\cos[(\omega_n + \omega_a)t - (\alpha_n + \alpha_a) - \tan^{-1}(\omega_n + \omega_a)\tau]}{[1 + (\omega_n + \omega_a)^2 \tau^2]^{1/2}} \right\}. \end{aligned} \quad (\text{III.27})$$

We note that if the range of optical frequencies is small compared with the average frequency, we can neglect the second group of terms compared with the first. Thus,

$$\begin{aligned} \delta n = & \frac{Dc\tau}{2\pi} \sum_{n,a} k_n k_a |A_n| |A_a| \\ & \times \frac{\cos[(\omega_n - \omega_a)t - (\alpha_n - \alpha_a) - \tan^{-1}(\omega_n - \omega_a)\tau]}{[1 + (\omega_n - \omega_a)^2 \tau^2]^{1/2}}. \end{aligned} \quad (\text{III.28})$$

This result, being based on the invalid conjecture of Eq. (III.24), is now on a very tenuous footing. In the following section, we use a different derivation which will arrive at the same result, putting it on a much firmer basis.

#### IV. MODIFIED SEMICLASSICAL TREATMENT OF RADIATION

In order to eliminate the problems we found in treating coherent radiation in the usual semiclassical manner,<sup>12</sup> we propose a modification which retains the essential features of the classical treatment of the field and quantum treatment of the electrons but which takes account of the lifetime of excited states in the quantum-mechanical equations of motion for these states.

Equation (III.3) assumes that if the radiation field is turned off, the electron no longer makes any transition, whereas we know that it relaxes from the excited state back to the normal state. For the case in which the relaxation is exponential with a time constant  $\tau$ , the relaxation mechanism is accounted for in a phenomenological way by adding an additional term  $H_1$  to the Hamiltonian. We take this additional term to be diagonal in the eigenstates of the unperturbed Hamiltonian, with diagonal matrix elements

$$(H_1)_{jj} = -i\hbar/2\tau \quad (\text{IV.1})$$

for any excited state  $j$ ; all other matrix elements are zero. The equation of motion (Eq. III.3) then becomes

<sup>12</sup> See Ref. 2. This method is reminiscent of the Weisskopf-Wigner method. See W. Shockley, *Electrons and Holes in Semiconductors* (D. Van Nostrand Company, Inc., New York, 1950), p. 487 ff.

(for any excited state  $j$ )

$$\dot{a}_j = (-i/\hbar) \sum_m a_m e^{i\omega_m t} \mathcal{C}_{jm} - (a_j/2\tau). \quad (IV.2)$$

We see that if the radiation is turned off at time  $t_0$ , then

$$a_j(t) = a_j(t_0) e^{-(t-t_0)/2\tau}, \quad (IV.3)$$

and thus

$$P_j(t) = |a_j(t)|^2 = P_j(t_0) e^{-(t-t_0)/\tau}, \quad (IV.4)$$

which indeed exhibits the desired exponential decrease in the probability that the electron is in an excited state, the time constant being  $\tau$ .

Taking account of the lifetime in this manner assures us throughout that the probability that the electron is in an excited state remains small and the probability that it is in the normal state remains close to unity for all time. The fact that the matrix elements of  $H_1$  are imaginary simply reflects the fact that we are not conserving probability (i.e., we are not accurately counting the occupancy of the normal state) when we approximate the probability of occupancy of the normal state by unity. Note also that we do not trace the actual return path followed by an electron between the state to which it was excited and the normal state. For example, in an  $n$ -type extrinsic photoconductor, we do not trace the actual transitions an electron might make within the conduction band before returning to a donor impurity state, since the photocurrent is essentially independent of the exact position of the electron in the conduction band.

Assuming again the same initial conditions as in Sec. III, i.e.,

$$a_m(0) = \delta_{mI}, \quad (IV.5)$$

the recombination process now assures us that for all time

$$a_I(t) \approx 1 \quad \text{and} \quad a_j(t) \ll 1, \quad j \neq I, \quad (IV.6)$$

and hence, from Eq. (IV.2), we find to first order

$$\dot{a}_{FI} = (-i/\hbar) e^{i\omega_{FI} t} \mathcal{C}_{FI} - (a_{FI}/2\tau). \quad (IV.7)$$

Since the above equation of motion, unlike Eq. (III.5), is valid for all time, we can solve directly for the steady state excitation probability rather than just the excitation rate. The steady-state solution is

$$a_{FI}(t) = \frac{2\tau}{i\hbar} \sum_n \sum_{m=1}^2 \mathcal{C}_{FI}^{nm} \frac{e^{i[\omega_{FI} + (-)^m \omega_n] t}}{1 + i[\omega_{FI} + (-)^m \omega_n] 2\tau}. \quad (IV.8)$$

Summing over initial and final states, and replacing the sum over initial states by an integral, we find

$$P(t) = \frac{4\tau^2}{\hbar^2} \sum_F \int_{-\infty}^{\infty} d\omega_I \rho(\omega_I) \times \sum_{n,a} \sum_{m,b=1}^2 \mathcal{C}_{FI}^{nm} (\mathcal{C}_{FI}^{ab})^* V_{FI}^{nm,ab}, \quad (IV.9)$$

where

$$V = e^{i2\theta t} / \{ [1 + i(\Omega + \theta)2\tau] [1 - i(\Omega - \theta)2\tau] \}, \quad (IV.10)$$

$\Omega$  and  $\theta$  are given by Eq. (III.12). Thus

$$P(t) = \frac{4\tau^2 e^2}{m^2 c^2} \sum_{F,n,a} \int_{-\infty}^{\infty} d\omega_I \rho(\omega_I) \times \left\{ \frac{A_n(\beta_n)_{FI} A_a^* [(\beta_a)_{FI}]^* e^{i(\omega_a - \omega_n)t}}{[1 + i2\tau(\omega_{FI} - \omega_n)] [1 - i2\tau(\omega_{FI} - \omega_a)]} + \frac{A_n^*(\beta_n^*)_{FI} A_a [(\beta_a^*)_{FI}]^* e^{i(\omega_n - \omega_a)t}}{[1 + i2\tau(\omega_{FI} + \omega_n)] [1 - i2\tau(\omega_{FI} + \omega_a)]} + \frac{A_n(\beta_n)_{FI} A_a [(\beta_a^*)_{FI}]^* e^{-i(\omega_n + \omega_a)t}}{[1 + i2\tau(\omega_{FI} - \omega_n)] [1 - i2\tau(\omega_{FI} + \omega_a)]} + \frac{A_n^*(\beta_n^*)_{FI} A_a^* [(\beta_a)_{FI}]^* e^{i(\omega_n + \omega_a)t}}{[1 + i2\tau(\omega_{FI} + \omega_n)] [1 - i2\tau(\omega_{FI} - \omega_a)]} \right\}. \quad (IV.11)$$

Note that the magnitude of  $[1 \pm i(\Omega \pm \theta)2\tau]^{-1}$  has a peak at  $\Omega = \mp \theta$  with half-width  $\sqrt{3}\tau/2$ .  $V$  is the product of two such peaked functions. If  $\theta \lesssim 1/\tau$ , then the peaks overlap, and  $V$  is largest around  $\Omega = 0$ . If we assume that the matrix elements and density of states in Eq. (IV.11) vary slowly over a range of frequencies of the order of  $1/\tau$ , we can then take them outside the integral (evaluating at  $\Omega = 0$ ), provided  $\theta \lesssim 1/\tau$ . Some terms in Eq. (IV.11) have values of  $\theta$  of the order of optical frequencies, i.e., of the order of  $10^{14} \text{ sec}^{-1}$  and since lifetimes in photoconductors are typically much longer than  $10^{-9} \text{ sec}$ , we have  $\theta \sim 10^5/\tau \gg 1/\tau$ . For these terms  $V$  is the product of two peaked functions whose peaks are widely separated, and thus these terms are small compared to those for which  $\theta \lesssim 1/\tau$ . They will be neglected henceforth. Again, because of the Pauli exclusion principle, the number of allowed final states is negligible for  $\omega_F < \omega_I$ . Thus, we find that Eq. (IV.11) simplifies to

$$P(t) = \frac{e^2}{m^2 c^2} \sum_{F,n,a} \rho(\omega_I) A_n A_a^* (\beta_n)_{FI} \times [(\beta_a)_{FI}]^* e^{i(\omega_a - \omega_n)t} \Big|_{\omega_{FI} = (\omega_n + \omega_a)/2} \times \int_{-\infty}^{\infty} \left[ \Omega_{FI}^{n1,a1} + \left( \theta^{n1,a1} - \frac{i}{2\tau} \right) \right]^{-1} \times \left[ \Omega_{FI}^{n1,a1} - \left( \theta^{n1,a1} - \frac{i}{2\tau} \right) \right]^{-1} d\Omega_{FI}^{n1,a1}. \quad (IV.12)$$

The remaining integral is easily evaluated by a contour integration. The result is

$$P(t) = \frac{2\pi\tau e^2}{m^2 c^2} \sum_{F,n,a} \rho(\omega_I) |A_n A_a (\beta_n)_{FI} (\beta_a)_{FI}| \times \frac{\cos[(\omega_n - \omega_a)t - (\alpha_n - \alpha_a) - \tan^{-1}(\omega_n - \omega_a)\tau]}{[1 + (\omega_n - \omega_a)^2 \tau^2]^{1/2}}, \quad (IV.13)$$

$\omega_{FI} = (\omega_a + \omega_n)/2.$

If the range of incident frequencies is small compared with the average frequency, then the probable number of excited electrons which we find from Eq. (IV.13) agrees with that found in Eq. (III.28).

Let us compare the modified derivation with the semiclassical one. In our modified treatment, we found a function  $V$  which is not really a delta function, and thus does not accurately conserve energy. However, the spread of this peak is caused by the finite lifetime  $\tau$ , as would be expected from uncertainty principle considerations, and decreases inversely with the lifetime. In the semiclassical derivation, we found a function  $W$  which had a delta-like part and a nondelta-like part  $g(\Omega, \theta, t)$ . The delta-like part is spread out with a width of order  $1/t$ , and since the treatment is valid only for times short compared with the relaxation time, the spreading is greater than  $1/\tau$ . The function  $g(\Omega, \theta, t)$  was simply discarded to obtain Eq. (III.28). Omission of this term was a procedure we could not justify. The physical meaning of this neglected term is not clear.

Thus, we see that in our modified treatment the approximation of  $V$  by delta functions is at least as good as the approximation of  $\sin xt/x$  by a delta function in the semiclassical treatment. Also, the deviation of  $V$  from a delta function is easily understood physically, whereas that of  $W$  is not clear.

## V. SUMMARY

Our theoretical study of absorption of light has demonstrated the difficulties which arise in an attempt to apply the usual semiclassical treatment to coherent radiation. These difficulties were eliminated in a modified treatment which considered (phenomenologically) the finite lifetimes of excited states in the quantum-mechanical equations of motion.

The result found in Eq. (IV.13) suggests that the lifetime would be determined by measurement of the frequency response of the photosignal to the beat frequency between two plane waves. This experimental technique is discussed by Penchina and Levinstein (see Ref. 1), who find good agreement between experiment and the theoretical result of Eq. (IV.13).

The same theoretical result can be derived in a completely quantum-mechanical treatment using the density matrix (Penchina, Quantum Theory of Optical Beating in Photoconductors, Ref. 2). Although more rigorous, that treatment lacks the intuitive simplicity of the modified semiclassical treatment of this paper.

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## APPENDIX

### A. Conjecture of Eq. (III.24)

We evaluate the integral of Eq. (III.24) explicitly for the simple case where

$$\begin{aligned} f(\Omega) &= 1, \quad \theta + A \leq \Omega \leq \theta + B, \quad B > A > 0 \\ f(\Omega) &= 0 \quad \text{for all other values of } \Omega. \end{aligned} \quad (\text{A1})$$

We find

$$\begin{aligned} & \int_{-\infty}^{\infty} f(\Omega) g(\Omega, \theta, t) d\Omega \\ &= \int_{\theta+A}^{\theta+B} g(\Omega, \theta, t) d\Omega \\ &= 2i \left( \theta \int_{\theta+A}^{\theta+B} \frac{d\Omega}{\Omega^2 - \theta^2} - \theta \cos \theta t \int_{\theta+A}^{\theta+B} \frac{\cos \Omega t d\Omega}{\Omega^2 - \theta^2} \right. \\ & \quad \left. - \sin \theta t \int_{\theta+A}^{\theta+B} \frac{\Omega \sin \Omega t}{\Omega^2 - \theta^2} d\Omega \right). \end{aligned} \quad (\text{A2})$$

The second and third terms above decrease rapidly due to cancellation as  $t$  increases. Thus for sufficiently large times we get

$$\begin{aligned} \int_{-\infty}^{\infty} f(\Omega) g(\Omega, \theta, t) d\Omega &\approx 2i\theta \int_{\theta+A}^{\theta+B} \frac{d\Omega}{\Omega^2 - \theta^2} \\ &= i \ln \frac{B(2\theta+A)}{A(2\theta+B)} \neq 0, \end{aligned} \quad (\text{A3})$$

which can indeed become quite large if  $A \ll B$ ,  $\theta$ . Hence we have disproved the conjecture of Eq. (III.24).

### B. Semiclassical Generation Rate

From Eq. (III.20), if we neglect  $g(\Omega, \theta, t)$  in Eq. (III.22), we find for  $t$  sufficiently large

$$\frac{dP_{FI\text{interference}}}{dt} \approx \frac{\pi}{\hbar^2} \sum'_{nm, ab} \mathcal{C}_{FI}{}^{nm} (\mathcal{C}_{FI}{}^{ab})^* e^{i2\theta t} [\delta(\Omega - \theta) + \delta(\Omega + \theta)], \quad (\text{A4})$$

$$\begin{aligned}
\frac{dP_{\text{interference}}}{dt} &= \sum_F \int_{-\infty}^{\infty} \rho(\omega_I) \frac{dP_{\text{FIinterference}}}{dt} d\omega_I \\
&\approx \sum_F \frac{\pi}{\hbar^2} \sum'_{nm, ab} (\mathcal{J}C_{FI}^{nm}) (\mathcal{J}C_{FI}^{ab})^* e^{i2\theta t} \rho(\omega_I) |_{\Omega=\theta} + |_{\Omega=-\theta} \\
&= \frac{\pi e^2}{m^2 c^2} \sum_F \left\{ \sum_{n, a; n \neq a} A_n(\beta_n)_{FI} A_a^* [(\beta_a)_{FI}]^* e^{i(\omega_n - \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=\omega_a} + |_{\omega_{FI}=\omega_n} \right. \\
&\quad + \sum_{n, a; n \neq a} A_n^*(\beta_n^*)_{FI} A_a [(\beta_a^*)_{FI}]^* e^{i(\omega_n - \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=-\omega_a} + |_{\omega_{FI}=-\omega_n} \\
&\quad + \sum_{n, a} A_n(\beta_n)_{FI} A_a [(\beta_a^*)_{FI}]^* e^{-i(\omega_n + \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=\omega_n} + |_{\omega_{FI}=-\omega_a} \\
&\quad \left. + \sum_{n, a} A_n^*(\beta_n^*)_{FI} A_a^* [(\beta_a)_{FI}]^* e^{i(\omega_n + \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=\omega_a} + |_{\omega_{FI}=-\omega_n} \right\}, \quad (\text{A5})
\end{aligned}$$

where we have used Eqs. (III.8) and (III.10). In Eq. (III.8) we replaced  $\sum_I$  by  $\int d\omega_I \rho(\omega_I)$ .

We note that the sum over allowed final states vanishes due to the Pauli exclusion principle if  $\omega_{FI} < 0$ . Also we note that  $n$  and  $a$  above are dummy summation indices. Thus

$$\begin{aligned}
\frac{dP_{\text{int}}}{dt} &= \frac{\pi e^2}{m^2 c^2} \sum_F \left\{ \sum_{n, a; n \neq a} A_n(\beta_n)_{FI} A_a^* [(\beta_a)_{FI}]^* \right. \\
&\quad \times e^{-i(\omega_n - \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=\omega_n} + \text{c.c.} \\
&\quad + \sum_{n, a} A_n(\beta_n)_{FI} A_a [(\beta_a^*)_{FI}]^* \\
&\quad \left. \times e^{-i(\omega_n + \omega_a)t} \rho(\omega_I) |_{\omega_{FI}=\omega_n} + \text{c.c.} \right\}. \quad (\text{A6})
\end{aligned}$$

Combining this with  $dP_{\text{direct}}/dt$  from Eq. (III.32), we find

$$\begin{aligned}
\frac{dP}{dt} &= \frac{2\pi e^2}{m^2 c^2} \sum_{F, n, a} \text{Re} A_n(\beta_n)_{FI} \\
&\quad \times \{ A_a^* [(\beta_a)_{FI}]^* e^{-i(\omega_n - \omega_a)t} \rho(\omega_I) \\
&\quad + A_a [(\beta_a^*)_{FI}]^* e^{-i(\omega_n + \omega_a)t} \rho(\omega_I) \}_{\omega_{FI}=\omega_n}, \quad (\text{A7})
\end{aligned}$$

where  $\text{Re} \equiv$  the real part of what follows.

If the solid has a center of inversion (e.g., perfect crystal of germanium, or one in which impurities are uniformly distributed), then the stationary states  $u_F$  and  $u_I$  have definite parity under inversion and we then

see from Eq. (III.10) that

$$(\beta_a^*)_{FI} = -(\beta_a)_{FI} \quad \text{for a solid with inversion symmetry.} \quad (\text{A8})$$

Then Eq. (7) becomes

$$\begin{aligned}
\frac{dP}{dt} &= \frac{2\pi e^2}{m^2 c^2} \sum_{F, n, a} \text{Re} A_n(\beta_n)_{FI} \rho(\omega_I) \\
&\quad \times \{ A_a^* [(\beta_a)_{FI}]^* e^{-i(\omega_n - \omega_a)t} \\
&\quad - A_a [(\beta_a)_{FI}]^* e^{-i(\omega_n + \omega_a)t} \}_{\omega_{FI}=\omega_n}. \quad (\text{A7a})
\end{aligned}$$

Let

$$A_j = |A_j| e^{i\alpha_j}, \quad (\beta_j)_{FI} = |(\beta_j)_{FI}| e^{i\gamma_j \mathbf{F} \cdot \mathbf{i}}, \quad (\text{A9})$$

then

$$\begin{aligned}
\frac{dP}{dt} &= \frac{2\pi e^2}{m^2 c^2} \sum_{F, n, a} |A_n A_a (\beta_n)_{FI} (\beta_a)_{FI}| \rho(\omega_I) \\
&\quad \times \{ \cos[(\omega_n - \omega_a)t - (\alpha_n - \alpha_a) - (\gamma_n \mathbf{F} \cdot \mathbf{i} - \gamma_a \mathbf{F} \cdot \mathbf{i})] \\
&\quad - \cos[(\omega_n + \omega_a)t - (\alpha_n + \alpha_a) - (\gamma_n \mathbf{F} \cdot \mathbf{i} - \gamma_a \mathbf{F} \cdot \mathbf{i})] \}_{\omega_{FI}=\omega_n}. \quad (\text{A10})
\end{aligned}$$

If the range of light frequencies is small so that all  $|(\beta_n)_{FI}| \approx |(\beta_N)_{FI}|$ ,  $\gamma_n \mathbf{F} \cdot \mathbf{i} \approx \gamma_N \mathbf{F} \cdot \mathbf{i}$ ,  $k_n \approx k_N$ , then Eq. (III.26) is verified. Note that if the solid does not have a center of symmetry, Eq. (III.26) no longer is valid. Still, Eq. (III.28) does follow as the only change is in the high-frequency terms which are neglected in any case.