field strength and rotating field is indicated in Fig. 4.

It should be pointed out that the width of a single coil resonance depends quite strongly on the form of the deflectability factor $f(\Delta m_J/J)$, particularly for small $\Delta m_J/J$, since this determines the contribution made by small rotations such as are obtained far from resonance. This factor in turn depends greatly on the slit arrangement. For instance, if there is a finite source slit and the detector slit is wide enough to just include the base of the resulting trapezoidal beam intensity cross section, the factor goes as $(\Delta m_J/J)^2$ rather than as $(\Delta m_J/J)$ for small $\Delta m_J/J$. This would give a narrower line than is indicated by the calculation here.

VII. COMPARISON WITH EXPERIMENT

The computer program has also been used to calculate the line shape corresponding to the conditions of an experimental line observed for the molecule OCS. This molecule is well suited for the purpose for several reasons; it consists of atoms whose nuclei are all spinless, so that there are no internal interactions to split the rotational magnetic-moment line; its relatively high moment of inertia means that at room temperature the most probable angular momentum J is 22, so that the classical limit should be applicable; and its easy handling and the occurrence of its mass at a low background of the mass spectrometer means a strong, quiet beam signal could be obtained. Multiple slits[2] were used to give high beam intensity for very narrow slitwidths. Experimental curves of the line shape were obtained for separated coils, both in phase and out of phase. These are shown in Fig. 5, together with the corresponding curves computed for the same conditions, including a deflectability factor chosen in accordance with the multiple slits.

The agreement between the maxima and minima of the interference patterns is very good. The fact that the experimental curves drop to zero much faster off-resonance than do the theoretical is probably due to the fringing field of the coil, so that the condition of non-adiabaticity does not hold, and the component of angular momentum follows the field and does not change. Inhomogeneities of the field H_0 are responsible for preventing the out-of-phase curve from dropping completely to zero at resonance.

The general agreement of the theoretical calculation with experiment confirms the applicability of the classical treatment of the multiple quantum transitions. The results of Sec. II offer a simple conception of the transition process which, for a full quantum mechanical treatment, would be extremely complicated. The fact that such transitions do occur makes possible the direct measurement of extremely small gyromagnetic ratios.

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Interaction of Very Intense Radiation Fields with Atomic Systems*

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In this paper a theory is developed which permits one to treat radiation processes involving a large number of photons in first- or second-order perturbation theory. The theory is applied to the interaction of an atomic electron with a very intense linearly polarized laser beam. It is found under certain approximations that induced radiation will occur at all harmonics $n\omega_0$ of the fundamental laser frequency ω_0. The intensity distribution of this radiation is symmetric about the axis of polarization of the primary beam and is peaked at an angle of about 45° with respect to direction of propagation of the secondary radiation and the polarization of the incident radiation for the first few modes. This angle markedly shifts toward 0° for increasing n (higher harmonics). The transition probabilities are high enough to make the effect readily observable.

I. INTRODUCTION

THEORETICAL work on the interaction of intense radiation (intensity $I > 10^5$ W/cm^2) with matter has recently received much stimulus with the advent of infrared and optical masers. Most detailed calculations have dealt with the radiation field in the classical approximation[1]; only a few approached the problem via quantum electrodynamics (QED).[2] Obviously the application of QED to a multiphoton problem is

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[1] See, for instance, K. Shimoda, T. C. Wang, and C. H. Townes, Phys. Rev. **102**, 1308 (1956); D. Kleppner, H. M. Goldenberg, and N. F. Ramsey, *ibid.* **126**, 603 (1962); Yoh Han Pao, J. Opt. Soc. Am. **52**, 871 (1962).

[2] Z. Fried, Phys. Letters **3**, 349 (1963); L. S. Brown and T. W. Kibble, Phys. Rev. **133**, A705 (1964). See also H. Paul, Ann. Physik **11**, 411 (1963); Z. Fried and J. H. Eberly, Bull. Am. Phys. Soc. **8**, 615 (1963); M. Mizushima, Phys. Rev. **132**, 951 (1963).

somewhat difficult since the QED is inherently geared to perturbation theory. A nonperturbative approximation is, of course, much more advisable. Recently, Fried[2] employed such a method, namely the well-known Bloch-Nordsieck method, for a calculation of Thomson scattering of intense radiation. But the method is unfortunately only applicable to free electrons. Here we propose an alternative approach which applies in principle also to bound states. This approach simply consists of considering $\mathbf{j}\cdot\mathbf{A}-\langle\mathbf{j}\cdot\mathbf{A}\rangle$ rather than $\mathbf{j}\cdot\mathbf{A}$ alone as a perturbation. $\mathbf{j}\cdot\mathbf{A}$ is of course the product of electron current and vector potential, the usual electron-photon interaction (we are using the Coulomb gauge), whereas $\langle\mathbf{j}\cdot\mathbf{A}\rangle$ is the expectation value of the interaction with respect to the photon variables alone. It therefore is still an operator in the electron variables. If certain modes of radiation are occupied by a large number of photons, the expression $\mathbf{j}\cdot\mathbf{A}-\langle\mathbf{j}\cdot\mathbf{A}\rangle$ is significantly different for these modes from $\mathbf{j}\cdot\mathbf{A}$ alone, whereas for modes which are unoccupied the difference between the two types of interaction is insignificant. $\mathbf{j}\cdot\mathbf{A}$ is a perturbation for a small number N of photons but ceases to be one if N increases, the expectation value being proportional to $\sqrt{N}$. On the other hand, $\mathbf{j}\cdot\mathbf{A}-\langle\mathbf{j}\cdot\mathbf{A}\rangle$ will always be a perturbation, the expectation value being zero. Subtracting from the interaction the expectation value $\langle\mathbf{j}\cdot\mathbf{A}\rangle$ of course modifies the Hamiltonian. In order to remedy the situation we add this term to the interaction-free Hamiltonian H_0 and consider $H_0+\langle\mathbf{j}\cdot\mathbf{A}\rangle$ as the new zero-order Hamiltonian with new modified eigenfunctions, etc., the starting point of a new perturbation calculation. If the photon number is large enough to warrant a transition to the classical limit for expectation values, then the new zero-order Hamiltonian is just the Hamiltonian of an electron in a given external electromagnetic field, and the new eigenfunctions are those corresponding to such a situation. So far, we did not mention the term proportional to A^2 in the nonrelativistic interaction Hamiltonian. Details will be given in Sec. III. Here it suffices to say that it will be treated completely analogously to the $\mathbf{j}\cdot\mathbf{A}$ term.

In Sec. II we will give preliminaries: the representation of the radiation fields to be used, transition to classical fields, decomposition of a laser beam in terms of eigenfunctions of the free radiation field, etc. In Sec. III the interaction theory will be developed along the ideas outlined above. Finally, in Sec. IV the theory will be applied to the induced emission of radiation from electrons subjected to a very intense laser beam ($I>10^{11}$ W/cm^2). The calculations are subject to certain approximations which will also be discussed.

II. PRELIMINARIES

It is convenient here and in the following to represent photons by their number and phase rather than by creation and destruction operators. For a photon of polarization α and propagation vector $\mathbf{k}$ the connection between these different sets of operators is[3]

$$a_\alpha^\dagger(\mathbf{k})=[N_\alpha(\mathbf{k})]^{1/2}e^{-i\phi_\alpha(\mathbf{k})},\quad a_\alpha(\mathbf{k})=e^{i\phi_\alpha(\mathbf{k})}[N_\alpha(\mathbf{k})]^{1/2}. \tag{1}$$

In a representation in which N is diagonal we have

$$\phi_\alpha(\mathbf{k})=-i[\partial/\partial N_\alpha(\mathbf{k})]. \tag{2}$$

The Hamiltonian of the free transverse radiation field is then given by

$$H_1=\hbar c\sum_{\alpha,\mathbf{k}} kN_\alpha(\mathbf{k}), \tag{3}$$

with eigenfunctions

$$\psi_{N'}=\prod_N \delta(N_\gamma(\mathbf{K})\,|\,N_\gamma'(\mathbf{K})), \tag{4}$$

where $\delta(N|N')$ is the Kronecker symbol and the function (4) is meant to be an infinite product over the countably infinite set of possible modes in the (large) quantization volume V. Obviously any wave function can be represented as a sum over the complete set (4):

$$\zeta(N_\gamma(\mathbf{K}))=\sum_{\text{all}\,N'}\zeta(N_\gamma'(\mathbf{K}))\prod_N \delta(N_\gamma(\mathbf{K})\,|\,N_\gamma'(\mathbf{K})). \tag{5}$$

The function φ is not necessarily a product of functions over all modes γ, $\mathbf{K}$ as in Eq. (4). It contains all the possible information about the radiation field. For instance,

$$|\zeta(N_\gamma(\mathbf{K}))|^2$$

is the probability to find $N_\gamma(\mathbf{K})$ photons of polarization γ and with propagation vector $\mathbf{K}$, etc. Therefore,

$$\sum_{\text{all}\,N}|\zeta(N_\gamma(\mathbf{K}))|^2=1, \tag{6}$$

is the proper normalization. Since the vector potential is given by

$$\mathbf{A}=\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\sum_{\alpha,\mathbf{k}}k^{-1/2}\boldsymbol{\varrho}_{\mathbf{k}}^{(\alpha)}\times\{e^{i(\mathbf{k}\cdot\mathbf{r}-ckt)}a_\alpha(\mathbf{k})+e^{-i(\mathbf{k}\cdot\mathbf{r}-ckt)}a_\alpha^\dagger(\mathbf{k})\}, \tag{7}$$

where the $\boldsymbol{\varrho}_k^{(\alpha)}$ are unit vectors of polarization, it is not difficult to find the expectation value of Eq. (7) with the help of Eqs. (1), (2), and (5):

$$\begin{aligned}\langle\zeta|\mathbf{A}|\zeta\rangle=\left(\frac{2\pi\hbar c}{V}\right)^{1/2}&\sum_{\alpha,\mathbf{k}}k^{-1/2}\boldsymbol{\varrho}_{\mathbf{k}}^{(\alpha)}\{e^{i(\mathbf{k}\cdot\mathbf{r}-ckt)}\\&\times\sum_{\text{all}\,N}\zeta^*(N_\gamma(\mathbf{K}))(N_\alpha(\mathbf{k})+1)^{1/2}\zeta(N_\gamma(\mathbf{K})\\&+\delta_{\alpha\gamma}\delta(\mathbf{K}|\mathbf{k}))+e^{-i(\mathbf{k}\cdot\mathbf{r}-ckt)}\sum_{\text{all}\,N}\zeta^*(N_\gamma(\mathbf{K})\\&\times(N_\alpha(\mathbf{k}))^{1/2}\zeta(N_\gamma(\mathbf{K})-\delta_{\alpha\gamma}\delta(\mathbf{K}|\mathbf{k}))\}.\end{aligned} \tag{8}$$

[3] W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, Oxford, England, 1957), 3rd ed., p. 65.

From

$$\mathbf{E}=-(1/c)(\partial\mathbf{A}/\partial t),\quad \mathbf{H}=\nabla\times\mathbf{A}, \tag{9}$$

expressions analogous to Eq. (8) may be obtained for the electric and magnetic fields.

It is often said that in order to be able to describe the electromagnetic fields classically the number of photons must be large. That this is not entirely true may readily be seen by taking the expectation value of the fields (9) with respect to the eigenfunction (4) of the Hamiltonian (3). No matter how large N' is, the expectation value is exactly zero. However, if the number of photons is not only large but uncertain, so that instead of using Eq. (4) we have to use Eq. (5), it is easy to show that the expectation value of any operator or any product of operators of the radiation field behaves as a classical quantity. To amplify this statement let us suppose that the wave function (5) is centered about a large value N^0 for certain modes. In other words, $\zeta(N)$ is only appreciable for N in the neighborhood of N^0 and negligibly small for other values of N. Also, for those modes of the radiation field which are not occupied, the wave function ζ consists of a product of Kronecker symbols $\delta(N|0)$. In this case the expectation value (8) of the vector potential may be written:

$$\langle\zeta|\mathbf{A}|\zeta\rangle=\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\sum_{\alpha,\mathbf{k}}{}' k^{-1/2}\boldsymbol{\varrho}_{\mathbf{k}}{}^{(\alpha)}2\cos(\mathbf{k}\cdot\mathbf{r}-ckt) \times\sum_{\text{all}N}|\zeta(N_\gamma(\mathbf{K}))|^2(N_\alpha(\mathbf{k}))^{1/2}. \tag{10}$$

The prime indicates that the sum over the modes α, $\mathbf{k}$ runs only over those modes which are occupied. To obtain Eq. (10) we have put

$$\zeta(N\pm1)\approx\zeta(N), \tag{11}$$

the error becoming vanishingly small with increasing N^0. From Eqs. (2) and (1) we also see that Eq. (11) is tantamount to assuming the operators a and $a^\dagger$ to be c numbers, for

$$\begin{aligned}a_\alpha(\mathbf{k})\zeta(N_\gamma(\mathbf{K}))&=\exp\{-[\partial/\partial N_\alpha(\mathbf{k})]\}(N_\alpha(\mathbf{k}))^{1/2}\zeta(N_\gamma(\mathbf{K}))\\&=(N_\alpha(\mathbf{k})+1)^{1/2}\zeta(N_\gamma(\mathbf{K})+\delta_{\gamma\alpha}\delta(\mathbf{K}|\mathbf{k}))\\&\approx(N_\alpha(\mathbf{k}))^{1/2}\zeta(N_\gamma(\mathbf{K})),\end{aligned} \tag{12}$$

using Eq. (11). It is also clear that the expectation value of a product of operators is equal to the product of the expectation values of the operators, provided that the approximation (11) holds. The expectation values of $\mathbf{E}$ and $\mathbf{H}$ are true classical fields.

We are now in a position to construct the wave function of a laser beam. For simplicity we assume a monochromatic plane-polarized beam. Let the set of photon modes γ, $\mathbf{K}$ be ordered in some way:

$$\gamma_1,\mathbf{K}_1;\cdots\quad\gamma_j,\mathbf{K}_j;\cdots\quad\gamma_i,\mathbf{K}_i;\cdots.$$

Let the laser beam have a center frequency corresponding to a wave vector $\mathbf{K}_j=\boldsymbol{\kappa}$ and polarization $\boldsymbol{\epsilon}$. We now put

$$\begin{aligned}\zeta(N_\gamma(\mathbf{K}))=&\,\delta(N_{\gamma_1}(\mathbf{K}_1)|0)\cdots\delta(N_{\gamma_{j-m}}(\mathbf{K}_{j-m})|0)\\&\times F(N_\epsilon(\mathbf{K}_{j-m+1}))\cdots F(N_\epsilon(\boldsymbol{\kappa}))\\&\times F(N_\epsilon(\mathbf{K}_{j+m-1}))\delta(N_{\gamma_{j+m}}(\mathbf{K}_{j+m})|0)\cdots,\end{aligned} \tag{13}$$

where

$$F(N)=(a/\pi)^{1/4}\exp\{-(a/2)(N^0-N)\}, \tag{14}$$

with large N^0. With the wave function (13) we then obtain for the electric field (the expectation value) taking the Eqs. (7) and (9) into account:

$$\langle\zeta|\mathbf{E}|\zeta\rangle=-2\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\sum_{\mathbf{k}}{}' k^{1/2}\boldsymbol{\varrho}_{\mathbf{k}}{}^{(\epsilon)}\sin(\mathbf{k}\cdot\mathbf{r}-ckt) \times\sum_N|F(N_\epsilon(\mathbf{k}))|^2[N_\epsilon(\mathbf{k})]^{1/2}. \tag{15}$$

Since N^0 in Eq. (14) is assumed to be very large, the sum over N in Eq. (15) is to a good approximation given by

$$\sum_N|F(N)|^2N^{1/2}=(N^0)^{1/2}. \tag{16}$$

It is now practical to go into the continuum, i.e., let the quantization volume go to infinity. In this case we have

$$V^{-1}\sum_{\mathbf{k}}\cdots\to(2\pi)^{-3}\int d^3k\cdots, \tag{17a}$$

$$N_\gamma(\mathbf{K})\to[(2\pi)^3/V]\bar{N}_\gamma(\mathbf{K}). \tag{17b}$$

With Eqs. (17) and (16) we obtain for the electric field:

$$\langle\zeta|\mathbf{E}|\zeta\rangle=-\pi^{-1}(\hbar c)^{1/2}\int d^3k\boldsymbol{\varrho}_{\mathbf{k}}{}^{(\epsilon)}k^{1/2} \times\sin(\mathbf{k}\cdot\mathbf{r}-ckt)[\bar{N}_\epsilon{}^0(\mathbf{k})]^{1/2}. \tag{18}$$

For future applications we wish Eq. (18) to represent a laser beam of frequency $\omega_0=c\kappa$ propagating into the Z direction of a space-fixed coordinate system with a polarization along the X axis (unit vector $\boldsymbol{\varrho}_x$). In order to do so we choose for $\bar{N}^0$.

$$\bar{N}^0=nb_1{}^{1/2}b_2\exp\{-b_1(k_z-\kappa)^2-b_2(k_x{}^2+k_y{}^2)\}. \tag{19}$$

The physical significance of the quantities $b_1{}^{1/2}$ and $b_2{}^{1/2}$ is easily seen through evaluation of the integral (18) with the expression (19). For large values of b_1 and b_2 we have, again to a very good approximation:

$$\langle\zeta|\mathbf{E}|\zeta\rangle=-2(2\pi\hbar\omega_0nb_1{}^{-1/2}b_2{}^{-1})^{1/2}\boldsymbol{\varrho}_x\sin(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega_0t) \times\exp\left\{-\frac{1}{2b_1}(Z-ct)^2-\frac{1}{2b_2}(x^2+y^2)\right\}. \tag{20}$$

This expression represents a laser pulse moving with velocity c in the Z direction. Clearly the quantities $b_1{}^{1/2}$ and $b_2{}^{1/2}$ signify the spatial extensions of the electric field, $b_2^{1/2}$ in x and y direction, $b_1^{1/2}$ in Z direction. For our purpose it is sufficient to consider a continuous laser beam, since the processes we will investigate are of short duration and are confined to a small volume (atomic system). In this case we let b_1 and b_2 go to infinity but such that the quantity $nb_1^{-1/2}b_2^{-1}$ stays finite. Putting

$$nb_1^{-1/2}b_2^{-1}=\tfrac{1}{2}\bar{n}\,, \tag{21}$$

which is obviously the number density of photons,[4] we have finally:

$$\langle\zeta\,|\,\mathbf{E}\,|\,\zeta\rangle=-(4\pi\hbar\omega_0\bar{n})^{1/2}\varrho_x\sin(\kappa Z-c\kappa t)\,. \tag{22}$$

The steps which lead from the state vector (13) of the photon field to the final expression for the expectation value of the electric field (22) may of course be applied to any other quantity of interest. Specificially, we will do this in the next section for the interaction Hamiltonian of an electron with the electromagnetic field. Details of the calculation will however be omitted. The reader is referred to this section for details.

III. DEVELOPMENT OF THE THEORY

The total nonrelativistic Hamiltonian of an electron in a given potential coupled to the radiation field is

$$H=H_0+H_1+H_i\,, \tag{23}$$

with

$$H_0=-(\hbar^2/2m)\nabla^2+\phi(\mathbf{r})\,, \tag{24}$$

the Hamiltonian of an electron in a given (attractive) potential,

$$H_1=\hbar c\sum_{\alpha,\mathbf{k}}kN_\alpha(\mathbf{k})\,, \tag{25}$$

the Hamiltonian of the free radiation field and

$$H_i=\frac{ie\hbar}{mc}\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\sum_{\alpha,\mathbf{k}}k^{-1/2}\varrho_{\mathbf{k}}^{(\alpha)}\cdot\nabla e^{i\mathbf{k}\cdot\mathbf{r}}(a_\alpha(\mathbf{k})+a_\alpha^\dagger(-\mathbf{k}))$$
$$\frac{\pi e^2\hbar}{mcV}\sum_{\alpha,\mathbf{k}}\sum_{\beta,\mathbf{k}'}(kk')^{1/2}\varrho_{\mathbf{k}}^{(\alpha)}\cdot\varrho_{\mathbf{k}'}^{(\beta)}e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}}$$
$$\times[a_\alpha(\mathbf{k})+a_\alpha^\dagger(-\mathbf{k})][a_\beta(\mathbf{k}')+a_\beta(-\mathbf{k}')]\,, \tag{26}$$

the interaction Hamiltonian. The creation and destruction operators for photons are represented by [see Eqs. (1) and (2)]

$$a_\alpha^\dagger(\mathbf{k})=[N_\alpha(\mathbf{k})]^{1/2}\exp\{-[\partial/\partial N_\alpha(\mathbf{k})]\}\,, \tag{27a}$$

$$a_\alpha(\mathbf{k})=\exp\{[\partial/\partial N_\alpha(\mathbf{k})]\}[N_\alpha(\mathbf{k})]^{1/2}\,. \tag{27b}$$

Let us assume that the atomic system (the electron) is subjected to an intense radiation field, the radiation field being represented by the state vector (13). We wish to separate a direct interaction with this radiation field, i.e., an interaction which does not change the number of photons, from an interaction which leads to a change in the number of photons. The latter, although being modified by the presence of the high-intensity radiation field, may still be evaluated by perturbation theory. Let us therefore try the following *ansatz* for the wave function of the electron and radiation field:

$$\psi=e^{-(it/\hbar)H_1}\zeta(N_\gamma(\mathbf{K}))\chi(\mathbf{r},t)\,. \tag{28}$$

Here ζ is given by Eq. (13). Inserting Eq. (28) into the time-dependent Schrödinger equation with Hamiltonian (23) and taking the expectation value with respect to the photon variables yields an equation for χ:

$$i\hbar(\partial/\partial t)\chi(\mathbf{r},t)=\{H_0+\langle\zeta\,|\,H_i(t)\,|\,\zeta\rangle\}\chi(\mathbf{r},t)\,. \tag{29}$$

In the derivation of Eq. (29) we have used the relationship

$$e^{(it/\hbar)H_1}\exp\left(\pm\frac{\partial}{\partial N_\alpha(\mathbf{k})}\right)e^{-(it/\hbar)H_1}$$
$$=e^{\mp ickt}\exp\left(\pm\frac{\partial}{\partial N_\alpha(\mathbf{k})}\right), \tag{30}$$

signifying that each term containing $\mathbf{k}\cdot\mathbf{r}$ in the interaction Hamiltonian (26) is simply replaced by $\mathbf{k}\cdot\mathbf{r}-ckt$. It is clear from Eq. (29) that the electronic wave function χ merely represents the electron being subjected to a given time-dependent external field.

We note here for later use that the expectation value of $H_i(t)$ occurring in Eq. (29) with the expression (13) for the wave function of the photons in the approximation discussed in Sec. II is given by

$$\langle\zeta\,|\,H_i(t)\,|\,\zeta\rangle=-\frac{e}{m}\left(\frac{4\pi\hbar\bar{n}}{\omega_0}\right)^{1/2}\cos(\kappa Z-\omega_0 t)\frac{\hbar}{i}\frac{\partial}{\partial x}$$
$$+\frac{2\pi\hbar e^2\bar{n}}{m\omega_0}\cos^2(\kappa Z-\omega_0 t)\,. \tag{31}$$

Since the Hamiltonian of Eq. (29) is Hermitian it is clear that the solutions of Eq. (29) form a complete orthonormal set of wave functions χ_n provided they were orthonormal at some initial time, which we assume to be the case henceforth. It is this set of wave functions which represents that part of the electron-photon interaction which does not disturb the number of photons. Let us now turn to the part of the interaction which does alter the number of photons. Consider the total Hamiltonian:

$$H=H_0+\langle\zeta\,|\,H_i(t)\,|\,\zeta\rangle+H_1+H_i-\langle\zeta\,|\,H_i(t)\,|\,\zeta\rangle\,, \tag{32}$$

which is of course identical to Eq. (23). But now we assume the last two terms on the right-hand side of Eq. (32) to be the perturbation. An expansion in terms of the eigenfunctions of Eq. (29) of the wave function ψ for the complete system satisfying the time-dependent Schrödinger equation with the Hamiltonian (32)

[4] The reason for the factor $\frac{1}{2}$ in Eq. (21) is simply that the mean time averaged energy density $(1/4\pi)\ (E^2+H^2)=\hbar\omega_0\bar{n}$.

leads to the following expression:

$$\psi=\sum_n f_n(N_\gamma(\mathbf{K}),t)\chi_n(\mathbf{r},t)e^{-(it/\hbar)H_1}. \tag{33}$$

Imposing the initial condition

$$f_n(N_\gamma(\mathbf{K}),0)=\delta_{nn'}\zeta(N_\gamma(\mathbf{K})), \tag{34}$$

where ζ is given by Eq. (13), first-order time-dependent perturbation theory then yields in a straightforward manner[5]:

$$f_n(N_\gamma(\mathbf{K}),t)=-\frac{i}{\hbar}\int_0^t d\tau\int d^3r\chi_n^*(\mathbf{r},\tau) \times[H_i(\tau)-\langle\zeta|H_i(\tau)|\zeta\rangle] \times\chi_{n'}(\mathbf{r},\tau)\zeta(N_\gamma(\mathbf{K})). \tag{35}$$

It is expression (35) which we will use in the next section to obtain induced emission probabilities for the system at hand.

IV. AN APPLICATION

Let us then apply Eq. (35) to a calculation of induced emission of radiation from an atomic electron. Physically we imagine that the laser beam is passing through a dilute gas. In this case we may ignore any correlations between different atoms either due to interactions among atoms or the coherence effects of the laser beam. We also imagine that due to the very high intensity of the incoming radiation all atoms are essentially ionized in a special way. To clarify this statement let us write down the Schrödinger equation for an outer electron of a given atom in the atomic field of force and the radiation field corresponding to Eq. (29) [using Eq. (31)]:

$$\left\{-\frac{\hbar^2}{2m}\nabla^2+\phi(\mathbf{r})+i\hbar c\alpha\cos(\kappa Z-\omega_0 t)\frac{\partial}{\partial x} +\tfrac{1}{2}(mc^2)\alpha^2\cos^2(\kappa Z-\omega_0 t)\right\}\chi_n=i\hbar(\partial/\partial t)\chi_n. \tag{36}$$

Here α is the dimensionless parameter

$$\alpha=(4\pi e^2\hbar\bar{n}/m^2c^2\omega_0)^{1/2} \tag{37}$$

measuring the strength of the radiation interaction. We now assume that the atomic potential is separable, i.e.,

$$\phi(\mathbf{r})=\phi_1(x)+\phi_2(y,z). \tag{38}$$

This approximation allows us to introduce two further approximations. First we can omit the z dependence in the cosine terms of Eq. (36) or, in other words, work in the dipole approximation, since the electron will stay bound in the y and z directions according to Eqs. (38) and (36). Second we may ignore the x-dependent part of the atomic force field entire, assuming very strong incident radiation. It is felt that the approximation (38) is, under the circumstances (namely: extremely strong linearly-polarized radiation, therefore a strong Stark effect in the x direction, little effect in the y and z directions) not at all a bad one.[6] Applying then the approximations outlined above to Eq. (36), it is not difficult to discover that the solution is given by

$$\chi_n=\psi_m(y,z)e^{-(it/\hbar)E_m}(2\pi)^{-3/2}\exp[iF_l(x,t)], \tag{39}$$

where

$$F_l(x,t)=lx-t((\hbar l^2/2m)+(mc^2/4\hbar)\alpha^2) +l(c/\omega_0)\alpha\sin\omega_0 t-(mc^2\alpha^2/8\hbar\omega_0)\sin 2\omega_0 t. \tag{40}$$

The wave function ψ_m is defined by

$$\left\{-\frac{\hbar^2}{2m}\left(\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial Z^2}\right)+\phi_2(y,Z)\right\}\psi_m=E_m\psi_m, \tag{41}$$

and the continuous wave function, $\exp(iF_l)$, satisfies

$$\left\{-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2}+i\hbar c\alpha\cos\omega_0 t\frac{\partial}{\partial x}+\frac{mc^2}{2}\alpha^2\cos^2\omega_0 t\right\}\exp[iF_l(x,t)] =i\hbar(\partial/\partial t)\exp[iF_l(x,t)], \tag{42}$$

an equation describing a one-dimensional free electron in a given time-dependent electric field. We see that indeed the wave function (39) satisfies Eq. (36) taking the above-mentioned approximations into account.

We are now in a position to work out the matrix element (35), since we know the wave functions χ_n from Eqs. (39) and (40) and we also know the photon state vector from Eqs. (13) and (14). In order to be consistent we have to use the same procedure in calculating the matrix element (35) as the one used in obtaining the expectation value (22) for the electric field, a procedure which has been thoroughly discussed in Sec. II. Proceeding then, we first notice that within the adopted approximations

$$\int d^3r\chi_n(\mathbf{r},\tau)\langle\zeta|H_i(\tau)|\zeta\rangle\chi_{n'}(\mathbf{r},\tau)=0, \tag{43}$$

provided that the quantum numbers of the initial state $n'\equiv m'$, l' are different from $n\equiv m$, l. This follows directly from the structure of the interaction term as given by the two last terms on the left-hand side of Eq. (42). Continuing with the evaluation of the matrix element (35) we specify the transition $n'\rightarrow n$ to be one in which a photon with polarization β and propagation vector $\mathbf{k}$ is emitted, whereas the electron undergoes a transition from the state specified by[7] $n'\equiv m$, l' to a state specified by $n\equiv m$, l. Inserting then the appropriate

[5] D. Bohm, *Quantum Theory* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1951), p. 410.

[6] Indeed under these circumstances the radiation field emerging from the sample will be that due to a harmonically oscillating free electron. We suspect that the quantum mechanical calculation will agree with a classical calculation as it is the case with Thomson scattering. Our result [Eq. (54)] will show this to be in fact true.

[7] m is the collection of quantum numbers associated with the motion in the y and z directions [Eq. (41)] and l is the (continuous) quantum number for the motion in x direction [Eq. (42)].

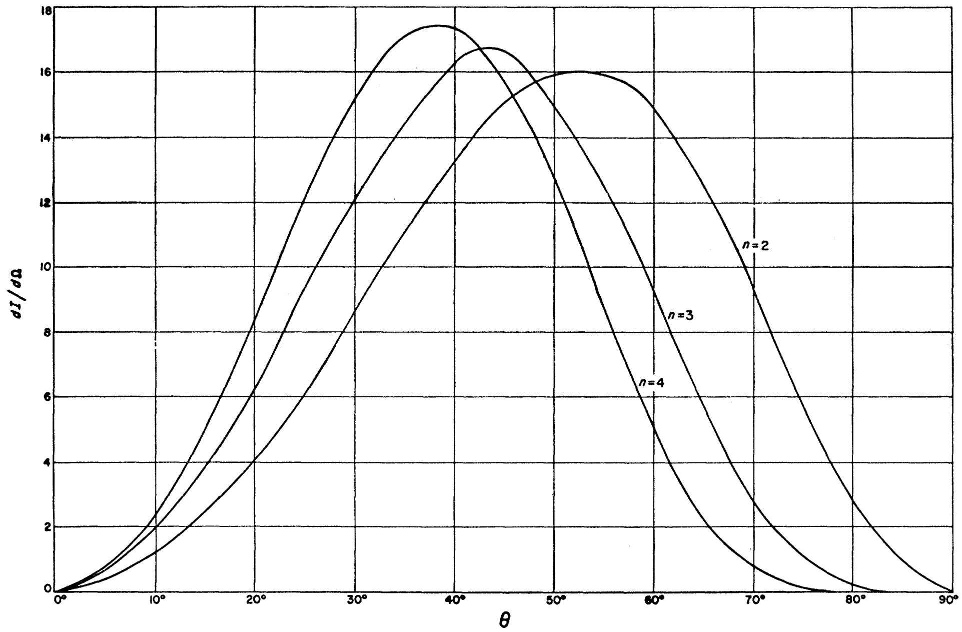

FIG. 1. Intensity distribution of secondary radiation in arbitrary units. The angle θ is the angle between the direction of polarization of the incident laser beam and the direction of propagation of the induced radiation. The distribution is shown for the first three harmonics and has been calculated with a value of α equal to one corresponding to a laser beam intensity of $I\approx 6.10^{13}$ Wcm^{-2} and a value for the electron velocity $v=2.10^8$ cm/sec.

wave functions (39) into the matrix element (35), performing the time integration, and selecting only the relevant part of the matrix element, i.e., that part which leads to a $\delta(N_\alpha(\mathbf{k})|1)$ from the expression (13) for the photon state, we obtain after some algebra for this amplitude A:

$$A=\frac{e}{mc}\left(\frac{2\pi\hbar c}{V}\right)^{1/2}\delta(l'-l-k_x)\boldsymbol{\varrho}_{\mathbf{k}}{}^{(\beta)}\cdot\boldsymbol{\varrho}_x l\sum_{n=-\infty}^{+\infty}J_n\left(\alpha\frac{l'-l}{\kappa}\right)\frac{\exp\{i[n\omega_0+ck+(\hbar/2m)(l^2-l'^2)]t\}-1}{n\omega_0+ck+(\hbar/2m)(l^2-l'^2)}\zeta(N_\gamma(\mathbf{K}))-\frac{2\pi e^2}{mcV}$$
$$\times\sum_{\delta,\mathbf{k}'}{}'(kk')^{-1/2}\boldsymbol{\varrho}_{\mathbf{k}}{}^{(\beta)}\cdot\boldsymbol{\varrho}_{\mathbf{k}'}{}^{(\delta)}\left\{\delta(l'-l-k_x+k_x')\sum_{n=-\infty}^{+\infty}J_n\left(\alpha\frac{l'-l}{\kappa}\right)\frac{\exp\{i[n\omega_0+c(k-k')+(\hbar/2m)(l^2-l'^2)]t\}-1}{n\omega_0+c(k-k')+(\hbar/2m)(l^2-l'^2)}\right.$$
$$\left.+\delta(l'-l-k_x-k_x')\sum_{n=-\infty}^{+\infty}J_n\left(\alpha\frac{l'-l}{\kappa}\right)\frac{\exp\{i[n\omega_0+c(k+k')+(\hbar/2m)(l^2-l'^2)]t\}-1}{n\omega_0+c(k+k')+(\hbar/2m)(l^2-l'^2)}\right\}[N_\delta(\mathbf{k}')]^{1/2}\zeta(N_\gamma(\mathbf{K})).\quad(44)$$

Although we do not wish to repeat here the algebraic steps which lead to Eq. (44), a few explanations are in order. The prime on the summation over δ and $\mathbf{k}'$ means that only those modes are to be summed which are occupied by the laser beam. α is the dimensionless quantity defined in Eq. (37). $\mathbf{k}$ is the propagation vector of the emitted photon, k_x of course its x component. The occurrence of the Bessel functions in expression (44) is due to the integration over time, since with wave functions of the type (39), (40) we encounter integrals of the type:

$$\int_0^t d\tau\,\exp(i\epsilon\tau+i\delta\sin\omega_0\tau)=-i\sum_{n=-\infty}^{+\infty}J_n(\delta)\frac{e^{it(\epsilon+n\omega_0)}-1}{\epsilon+n\omega_0},\quad(45)$$

a result which is readily verified using the generator function for Bessel functions $\exp[\frac{1}{2}(t-t^{-1})]$. Finally $\kappa=\omega_0/c$.

In order to find the transition probability per sec for the emission of a photon we have to take the absolute square of Eq. (44) and take the time derivative in the asymptotic limit $t\to\infty$.[8] It is clear from Eq. (44) that by taking the square two kinds of terms will be encountered. First (in easily understood notation) terms of the following kind:

$$\mathfrak{A}=\left|\frac{\exp\{it(n\omega_0+a+ck)\}-1}{n\omega_0+a+ck}\right|^2,\quad(46)$$

with the asymptotic limit for large times

$$\mathfrak{A}\to(2\pi/c)t\delta(k+c^{-1}(a+n\omega_0)),\quad(47)$$

[8] W. Heitler, Ref. 3, p. 139.

proportional to t leading to a constant transition probability in a well-known manner. But there are also other terms (cross terms) which are not as simply behaved. The cross terms

$$B=\frac{[\exp\{it(n\omega_0+a+ck)\}-1][\exp\{-it(m\omega_0+a'+ck)\}-1]}{(n\omega_0+a+ck)(m\omega_0+a'+ck)}+\text{complex conjugate}\,, \tag{48}$$

are oscillatory even in the asymptotic limit $t=\infty$. Specifically we have in this limit:

$$B\to\frac{2\pi}{c}\frac{\sin t[(n-m)\omega_0+a-a']}{(n-m)\omega_0+a-a'}[\delta(k+c^{-1}(n\omega_0+a))+\delta(k+c^{-1}(m\omega_0+a))]. \tag{49}$$

The transition probability will therefore consist of two parts: one part which is constant or time-independent, and another part which is oscillatory in time. Now, this time-dependent part is so rapidly oscillating, as can be seen from expression (49), that it is unobservable, its time average being zero. We may therefore omit all cross terms in performing the absolute square of Eq. (44). Multiplying also by the phase-space density of the photons, we obtain for the transition probability per solid angle and frequency interval dk:

$$\begin{aligned}\frac{d|A|^2}{dkd\Omega}=&\frac{e^2\hbar kl'^2}{4\pi m^2c^2}(\varrho_{\mathrm{k}}{}^{(\beta)}\cdot\varrho_x)^2\delta(l'-l-k_x)\sum_{n=-\infty}^{+\infty}\left[J_n\left(\frac{\alpha}{\kappa}(l'-l)\right)\right]^2\delta\left(k+c^{-1}\left[n\omega_0+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)+\frac{e^2k}{4\pi\hbar}\alpha^2(\varrho_{\mathrm{k}}{}^{(\beta)}\cdot\varrho_x)^2\\&\times\delta(l'-l-k_x)\left\{\sum_{n=-\infty}^{+\infty}\left[J_n\left(\frac{\alpha}{\kappa}(l'-l)\right)\right]^2\left[\delta\left(k+c^{-1}\left[(n-1)\omega_0+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)+\delta\left(k+c^{-1}\left[(n+1)\omega_0\right.\right.\right.\right.\\&\left.\left.\left.\left.+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)\right]+2\sum_{n=-\infty}^{+\infty}J_n\left(\frac{\alpha}{\kappa}(l'-l)\right)J_{n+2}\left(\frac{\alpha}{\kappa}(l'-l)\right)\delta\left(k+c^{-1}\left[(n+1)\omega_0+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)\right\}\\&-\frac{e^2\alpha l'k}{4\pi mc^2}(\varrho_{\mathrm{k}}{}^{(\beta)}\cdot\varrho_x)^2\delta(l'-l-k_x)\sum_{n=-\infty}^{+\infty}J_n\left(\frac{\alpha}{\kappa}(l'-l)\right)J_{n+1}\left(\frac{\alpha}{\kappa}(l'-l)\right)\\&\times\left\{\delta\left(k+c^{-1}\left[n\omega_0+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)+\delta\left(k+c^{-1}\left[(n+1)\omega_0+\frac{\hbar}{2m}(l^2-l'^2)\right]\right)^1\right\}.\end{aligned} \tag{50}$$

The total transition probability is found by summing over all final states. To be sure, we should also take an average over the initial states of the electron. But first we do not know their distribution and second we will see that the transition probabilities are rather insensitive to it. Summing over l in Eq. (50) and introducing the angle θ between the direction of propagation of the emitted photon and the x direction (the direction of polarization of the incident radiation), the arguments of the δ functions become

$$k+n\kappa+(\hbar/2mc)k^2\cos^2\theta-(\hbar/mc)l'k\cos\theta=0\,, \tag{51a}$$

or

$$k+(n\pm1)\kappa+(\hbar/2mc)k^2\cos^2\theta-(\hbar/mc)l'k\cos\theta=0\,. \tag{51b}$$

The roots of these equations correspond to a frequency of the emitted radiation ω given by

$$\omega=\frac{mc^2}{\hbar\cos^2\theta}\left\{-1+\left(1-2n\omega_0\frac{\hbar\cos^2\theta}{mc^2}\right)^{1/2}\right\}\approx-n\omega_0\,, \tag{52}$$

for not too large l'. We see that positive roots only occur for negative values of n, and the frequencies of the emitted radiation then are just the harmonics of the fundamental laser frequency ω_0. Neglecting recoil and introducing the initial velocity $v=hl'/m$ of the electron, we finally find for the differential transition probability of emitting a photon with frequency $n\omega_0$ and polarization β from Eq. (50):

$$\begin{aligned}\frac{d|A_\beta(n\omega_0)|^2}{d\Omega}=&\frac{n\omega_0}{4\pi}\frac{e^2}{\hbar c}(\varrho_{\mathrm{k}}{}^{(\beta)}\cdot\varrho_x)^2\left\{\frac{v^2}{c^2}[J_n(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x)]^2+\frac{v}{c}\alpha J_n(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x)(J_{n-1}(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x)+J_{n+1}(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x))\right.\\&\left.+\alpha^2(J_{n-1}(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x)+J_{n+1}(n\alpha\varrho_{\mathrm{k}}{}^{(1)}\cdot\varrho_x))^2\right\}.\end{aligned} \tag{53}$$

α is given by Eq. (37); $\boldsymbol{\varrho}_{\mathbf{k}}^{(1)}$ is a unit vector in direction of propagation of the emitted light.

Since the velocity of the electron v is according to Eq. (39) the average momentum divided by the mass and since the electron will be ejected from the atom with equal probability in either direction, we may omit the term linear in v in Eq. (53). Equation (53) then simplifies to:

$$\frac{d|A_\beta(n\omega_0)|^2}{d\Omega}=\frac{n\omega_0}{4\pi}\frac{e^2}{\hbar c}(\boldsymbol{\varrho}_{\mathbf{k}}^{(\beta)}\cdot\boldsymbol{\varrho}_x)^2[J_n(n\alpha\boldsymbol{\varrho}_{\mathbf{k}}^{(1)}\cdot\boldsymbol{\varrho}_x)]^2 \times\left(\frac{v^2}{c^2}+\frac{4}{(\boldsymbol{\varrho}_{\mathbf{k}}^{(1)}\cdot\boldsymbol{\varrho}_x)^2}\right). \quad (54)$$

This expression agrees with that given by Brown and Kibble[2] if magnetic interactions are omitted. It also agrees with the classical result for the radiation field of a harmonically oscillating point charge as given by Jackson[9] if v is put equal to zero. Since

$$\sum_\beta(\boldsymbol{\varrho}_{\mathbf{k}}^{(\beta)}\cdot\boldsymbol{\varrho}_x)^2=\sin^2\theta\,, \quad (55)$$

which vanishes for $\theta=0°$ and $180°$ and since the Bessel functions vanish when $\theta=90°$ or $270°$, we see that the secondary radiation field for each harmonic consists of four lobes rotationally symmetric about the axis of polarization of the primary laser beam. We notice that the direction of maximum intensity shifts toward 0° (i.e., toward the direction of polarization of the primary laser beam) with increasing frequency. Using the asymptotic expression for Bessel functions with large index and large argument it is not difficult to discover that the intensity distribution of the nth harmonic becomes for $\alpha<1$:

$$I_n\sim\frac{n\sin^2\theta}{\alpha^{-1}\cos\theta-\alpha\cos^2\theta}\exp\left\{-2n\left[\log\left(\frac{1+(1-\alpha^2\cos^2\theta)^{1/2}}{\alpha\cos\theta}\right)-\frac{1}{\alpha\cos\theta}+\alpha\cos\theta\right]\right\}. \quad (56)$$

[9] J. A. Jackson, *Classical Electrodynamics* (John Wiley & Sons, Inc., New York, 1962), p. 501.

This valid for large n but of course not too large n, both because eventually the recoil cannot be neglected any more, and because the nonrelativistic calculation becomes invalid. In any case Eq. (56) shows that the intensity is sharply peaked at an angle $\theta=(n\alpha/2)^{-1/2}$ rad. For an infrared laser operating at $\omega_0=10^{13}$ sec^{-1}, $n=100$ means ultraviolet light. But for $n=100$, expression (56) is perfectly valid, so that we expect two very sharp cones of ultraviolet radiation centered about the direction of polarization of the incident laser beam to emerge from our sample.

Turning now to the magnitude of the effect, we first notice that the transition probabilities (54) increase first with increasing intensity of the primary radiation. This is in contrast to the Thomson-scattering cross section, which is always inhibited with increasing intensity of primary radiation. As discussed by Fried[2] the reason for this decrease is just the opening up of more scattering channels with increasing intensity, those channels being precisely the ones we investigated here. On the other hand, if α goes to zero the transition probabilities (54) also vanish, since a free electron cannot spontaneously emit radiation. Of course the limit $\alpha=0$ in Eq. (54) is a mathematical limit, since the physical approximations which lead to Eq. (54) will break down long before α is zero. Assuming now for the electron velocity v a value of 2×10^8 cm/sec[10] numerical integration of Eq. (54) shows that for an infrared laser ($\omega_0=10^{13}$ sec^{-1}) the total transition probabilities are fairly large. Specificially we find a total transition probability of 10^8 sec^{-1} for the first few harmonics, assuming $\alpha=\frac{1}{5}$ corresponding to a laser beam intensity of 4.10^{12} W/cm^2.

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[10] This value is suggested from ionization experiments which show that the mean energy of the liberated (ionized) electrons is about 10 eV no matter what the cause of ionization. See H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, England, 1956).