The results of Madsen[8] and of Jesse and Sadauskis[9] for ionization in argon by recoils from alpha decay have been included in Fig. 4. The Madsen points have been corrected for the presence of 5% air in the argon gas. The results of the alpha recoil measurements seem to agree with the general trend of the present results.

W values for alpha particles having kinetic energies of several MeV have been observed[10] in argon and found to be independent of the particle energy within 0.5%. It is seen from Fig. 4 that for the heavier ions W is quite dependent on the particle velocity, increasing as the velocity decreases. It is also seen that for a given initial ion velocity, W increases as the mass of the ion increases, with the exception of argon ions at the lower velocities.

[8] B. S. Madsen, Kgl. Danske Videnskab. Selskab. Mat-Fys. Medd. **23**, No. 8 (1945).
[9] W. P. Jesse and J. Sadauskis, Phys. Rev. **102**, 389 (1956).
[10] W. P. Jesse, H. Forstat, and J. Sadauskis, Phys. Rev. **77**, 782 (1950).

ACKNOWLEDGMENTS

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Magnetic Resonance with Large Angular Momentum*

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Multiple quantum transitions present in molecular-beam magnetic resonance of molecules of high rotational angular momentum are treated in terms of a purely classical model. The probability of a change $(\Delta m_J/J)$ in rotational magnetic quantum number per unit angular momentum is shown to be given by $P(\Delta m_J/J)=1/[4\sin(\frac{1}{2}\epsilon)]$ if $|\Delta m_J/J|<2\sin(\frac{1}{2}\epsilon)$ and $P(\Delta m_J/J)=0$ if $|\Delta m_J/J|>2\sin(\frac{1}{2}\epsilon)$, where $P(\Delta m_J/J)\,d(\Delta m_J/J)$ is the fraction of molecules for which this change lies between $\Delta m_J/J$ and $\Delta m_J/J+d(\Delta m_J/J)$, and ϵ is the angle which the angular momentum makes with the magnetic field after the transition for a molecule whose angular momentum is initially along the field. By way of comparison, the transition probability of a spin-$\frac{1}{2}$ particle under the same conditions is $\sin^2(\frac{1}{2}\epsilon)$. This probability is weighted according to the probability of detecting a transition with a change in magnetic moment of $\mu_J\Delta m_J$ and averaged over the thermal distributions of J and V to give a theoretical line shape. The theory is applied to rotational magnetic moments of molecules. The calculated line shape is shown to agree reasonably well with an experimental curve of the rotational magnetic-moment resonance of the molecule OCS.

I. INTRODUCTION

RECENT work by Lawrence,[1] Anderson,[2] and Pinkerton[3] has indicated that in molecular-beam magnetic resonance of magnetic moments associated with large angular momenta an important role is played by multiple quantum transitions. The purpose of this paper is to present a completely classical treatment of these transitions, as should be valid in the limit of large angular momenta. The system considered consists of a molecule with a magnetic moment proportional to the angular momentum J, so that the gyromagnetic ratio g_J is independent both of J and its projection m_J along the field axis. This is very nearly true for all rotational moments. The molecule passes through a standard flop-out molecular-beam apparatus,[4] with an oscillating field perpendicular to a homogeneous magnetic field to produce a transition. A complete calculation has been made of the theoretical line shape, which agrees reasonably well with the experimental line shape of the rotational moment resonance of the molecule OCS. The values of the rotational moments measured in several molecules and the experimental techniques of measurement are discussed in another paper.[5]

II. SINGLE COIL DERIVATION

The system consists of a molecule with a magnetic moment $\mathbf{u}_J$ proportional to its angular momentum $\mathbf{J}\hbar$. The molecule passes through a rotating field inside of a homogeneous field region. The rotating field is given by

$$\mathbf{H}_1=H_1\cos\omega t\,\mathbf{i}-H_1\sin\omega t\,\mathbf{j},$$

* Work supported jointly by the National Science Foundation and the U. S. Navy Office of Naval Research.
[1] T. R. Lawrence, C. H. Anderson, and N. F. Ramsey, Phys. Rev. **130**, 1864 (1963).
[2] C. H. Anderson, Ph.D. thesis, Harvard University (1961) (unpublished).
[3] J. N. Pinkerton, Ph.D. thesis, Harvard University (1961) (unpublished).
[4] N. F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956).
[5] J. W. Cederberg, C. H. Anderson, and N. F. Ramsey (to be published).

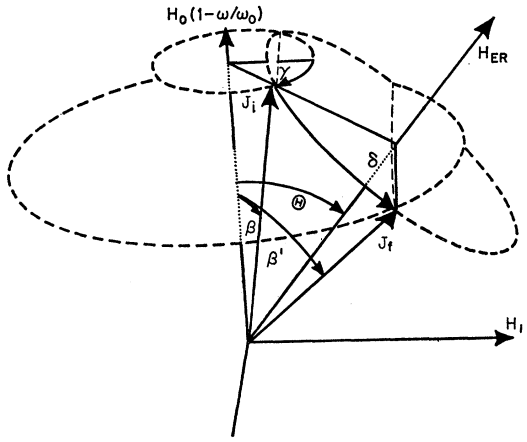

FIG. 1. Model of the classical magnetic moment transition. During the time the molecule is in the coil its angular momentum precesses from $\mathbf{J}_i$ to $\mathbf{J}_f$, changing its projection angle on H_0 from β to β'.

being perpendicular to the homogeneous field $H_0\mathbf{k}$. The analysis of the motion of the molecule is greatly simplified by the use of a coordinate system rotating with the field $\mathbf{H}_1$.[4]

In this system the effective field becomes

$$\begin{aligned}\mathbf{H}_{er}&=\mathbf{H}+\boldsymbol{\omega}H_0/\omega_0\\&=H_0(1-\omega/\omega_0)\mathbf{k}+H_1\mathbf{i},\end{aligned}\tag{1}$$

where $\omega_0=\mu_J/J\hbar H_0$ is the Larmor frequency of the molecule. Before the molecule enters the rotating field, H_1 is zero, and the angular momentum precesses about $\mathbf{H}_0$ with an angular velocity $\omega_0-\omega$. As it enters the coil it begins precessing about the resultant axis $\mathbf{H}_{er}$ with a frequency $\omega_0 H_{er}/H_0$. This process will be expressed in terms of the quantities $\theta=\cot^{-1}(\omega_0-\omega)/2b$ and $2b=\omega_0 H_1/H_0$, where θ (the angle between $\mathbf{H}_0$ and $\mathbf{H}_{er}$) and b indicate the frequency and the magnitude, respectively, of the rotating field as dimensionless variables.

If the coordinates describing the direction of the angular momentum at the instant it enters the coil are β, γ with respect to the axis of $\mathbf{H}_0$ as in Fig. 1, its precession about $\mathbf{H}_{er}$ will be through an angle

$$\begin{aligned}\delta&=\omega_0[(1-\omega/\omega_0)^2+(H_1/H_0)^2]^{1/2}t\\&=2b[\cot^2\theta+1]^{1/2}t\\&=(2b/\sin\theta)(l/V\alpha)\\&=B/\sin\theta V\end{aligned}\tag{2}$$

to a new direction given by β', γ'. Here l is the length of the coil, and $V\alpha$ is the velocity of the molecule with $\alpha=(2kT/m)^{1/2}$ and $B=2bl/\alpha$.

We wish to find the fraction of molecules with a given V and J which undergo a change $\Delta m_J/J$ (of projection m_J along the direction of H_0 per unit angular momentum) which lies between $\Delta m_J/J$ and $\Delta m_J/J+d(\Delta m_J/J)$. This fraction we will call $P(\Delta m_J/J)d(\Delta m_J/J)$. It will be shown by a simple geometrical argument that $P(\Delta m_J/J)$ is given by

$$\begin{aligned}P(\Delta m_J/J)&=1/[4\sin(\tfrac{1}{2}\epsilon)]\quad\text{for}\quad|\Delta m_J/J|<2\sin(\tfrac{1}{2}\epsilon)\\&=0\qquad\qquad\text{for}\quad|\Delta m_J/J|>2\sin(\tfrac{1}{2}\epsilon),\end{aligned}\tag{3}$$

where $\sin(\frac{1}{2}\epsilon)=\sin\theta|\sin(\frac{1}{2}\delta)|$, so that ϵ is the angle made finally with the H_0 axis by an angular momentum initially parallel to H_0. If the initial distribution of direction of the angular momentum is represented by a sphere of radius J, the fraction of the molecules which have their angular momentum directed within an area of the spherical surface is proportional to that area. The transition process is a rotation of the sphere about an axis at an angle θ with respect to the vertical axis, and the value of Δm_J for a point on the sphere is the distance the point moves up or down during the rotation. The quantity $P(\Delta m_J/J)\ d(\Delta m_J/J)$ is just the ratio of the area between the curves connecting the points for which vertical motion is Δm_J and $\Delta m_J+d\Delta m_J$ to the total area of the sphere.

By symmetry the points for which $\Delta m_J=0$ are those which lie in the plane which initially makes an angle $-\frac{1}{2}\delta$ with the plane of the vertical and the rotation axis, and which therefore makes a final angle $+\frac{1}{2}\delta$. Likewise, those points in a plane parallel to the first and at a distance h from it move vertically not by the plane's rotation in space, but only by its resultant translation. The vertical motion is the same for all points in the plane and may therefore be calculated for the point at the center of the circle of intersection with the sphere. This point moves a distance $2h\sin(\frac{1}{2}\delta)$ at an angle θ to the horizontal and hence moves vertically a distance $2h\sin(\frac{1}{2}\delta)\sin\theta$. Thus the two curves of Δm_J and $\Delta m_J+d\Delta m_J$ are just the intersections with the sphere of two parallel planes a distance $dh=d(\Delta m_J/J)/(2\sin\theta\sin\frac{1}{2}\delta)$ apart. The theorem of solid geometry that parallel planes cut off an area of a sphere which is independent of the height of the planes above the center gives immediately that $P(\Delta m_J/J)$ is independent of $\Delta m_J/J$. Furthermore the maximum value of Δm_J is that for which $|h|=J$, the radius of the sphere, or Δm_J max $=2J\sin\theta|\sin\frac{1}{2}\delta|$. Normalization then gives $P(\Delta m_J/J)=1/(4\sin\theta|\sin\frac{1}{2}\delta|)$.

It may be observed that $\sin\theta|\sin\frac{1}{2}\delta|$ is just $\sin\frac{1}{2}\epsilon$, where ϵ is the value of β' for a molecule with $\beta=0$, or, in terms of the sphere, it is the angle through which the point initially at the top rotates. The fact that the probability distribution depends only on this angle means that for more complicated situations, such as separated coils, only this angle need be computed. The quantity $\sin^2\frac{1}{2}\epsilon$ is also just the transition probability of a spin-$\frac{1}{2}$ particle of the same gyromagnetic ratio under the same conditions.[4]

III. COMPARISON WITH MAJORANA FORMULA

The Majorana formula gives the general quantum mechanical transition probability for a particle of spin

J in terms of ϵ as

$$P_{m_J,m_J'}=(J-m_J)!(J+m_J)!\times(J-m_J')!(J+m_J')!(\sin\tfrac{1}{2}\epsilon)^{4J}\times\left[\sum_r\frac{(-1)^r(\cot\tfrac{1}{2}\epsilon)^{m_J+m_J'+2r}}{(J-m_J-r)!(J-m_J'-r)!(m_J+m_J'+r)!r!}\right]^2. \tag{4}$$

What has been calculated here may be regarded as the limit, for high J, of

$$P(\Delta m_J,J)=\sum_{m_J}P_{m_J,m_J+\Delta m_J}. \tag{5}$$

In order to provide a comparison between the classical and quantum results, the lines for $P(\Delta m_J,J)$ are plotted in Fig. 2 for $J=2$, $\epsilon=\frac{1}{2}\pi$ and $\frac{1}{3}\pi$ together with the comparable classical curves. It indicates that even for this low value of J the quantum-mechanical distribution shows the approximate features of the classical one.

IV. DOUBLE COIL CALCULATION

In order to calculate $P(\Delta m_J/J)$ for two separated coils, which was the system used for this experiment, it is necessary to calculate the angle ϵ for the total change produced by, first, a precession about $\mathbf{H}_{er}$ through an angle δ, then a precession about $\mathbf{H}_0$ through an angle 2ν, and, finally, a precession again about $\mathbf{H}_{er}$ through an angle δ. The precession about $\mathbf{H}_0$ is at a rate $2b\cot\theta$ for a time $2lL/V\alpha$, where L is the ratio of the length of the gap between the coils to the total length of the two

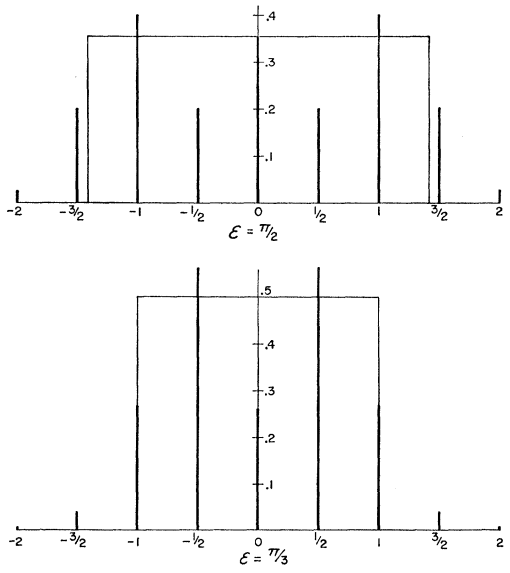

FIG. 2. Comparison with quantum mechanical Majorana formula for $J=2$. The rectangular curve is a plot of the function $P(\Delta m_J/J)$ calculated here, shown in relation to the corresponding quantum mechanical probabilities $J/(2J+1)\,\Sigma_m P_{m,m+\Delta m_J}$ (heavy vertical lines).

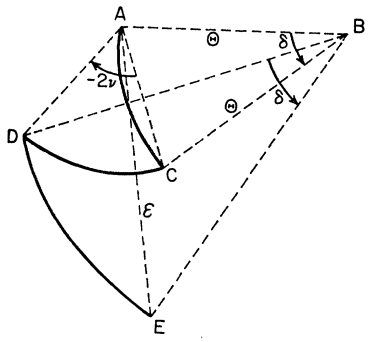

FIG. 3. Transition process for double coil. The angular momentum of the molecule, initially parallel to the field, takes the path $ACDE$ in reaching a final angle ϵ with the field. For simplicity, the spherical triangles are drawn as plane.

coils. If there is in addition a phase shift λ between the rotating fields of the two coils, this is equivalent to subtracting the phase shift angle from the angle of precession. The angle 2ν is therefore $2\nu=2BL\cot\theta/V-\lambda$. The diagram in Fig. 3 indicates this situation, with the simplification that the spherical triangles are drawn as plane.

Application of the spherical triangle relationships for ABC, ABD, and ABE gives the result

$$2\sin\tfrac{1}{2}\epsilon=2\sin\theta\,|\sin\delta\cos\nu-(1-\cos\delta)\cos\theta\sin\nu|\,. \tag{6}$$

V. DEFLECTABILITY WEIGHTING FACTOR

It has been shown that the fraction of the molecules with a fixed J and V which undergo a change of between $\Delta m_J/J$ and $\Delta m_J/J+d\Delta m_J/J$ is given by Eq. (1). The quantity $2\sin\frac{1}{2}\epsilon$ has been calculated for both the single and double coil cases. The signal produced by these molecules at the detector will be dependent on the relationship of the distance they are deflected to the slit dimensions. The displacement of a molecule from a refocused position at the detector is

$$S=\Delta\mu(\partial H/\partial z)(l_Bl_D)/(m\alpha^2V^2), \tag{7}$$

where $\Delta\mu=\hbar Jg_J\mu_N|\Delta m_J/J|$ is the effective change of magnetic moment, m is the mass of the molecule, $\partial H/\partial z$ is the gradient of the B magnet, $\alpha=(2kT/m)^{1/2}$ is the most probable velocity of the molecules in the source, l_B is the length of the B magnet, and l_D is the distance from the center of the B magnet to the detector slits. This may be written

$$S=W_1S_1J|\Delta m_J/J|/V^2, \tag{8}$$

where

$$S_1=(1/W_1)g_J\mu_N\hbar(\partial H/\partial z)(l_Bl_D)/(2kT). \tag{9}$$

For a single detector slit of width W_1 and an infinitely narrow source slit, the fraction of molecules deflected out of the refocused position will be proportional to the deflection up to a maximum of one, which occurs when

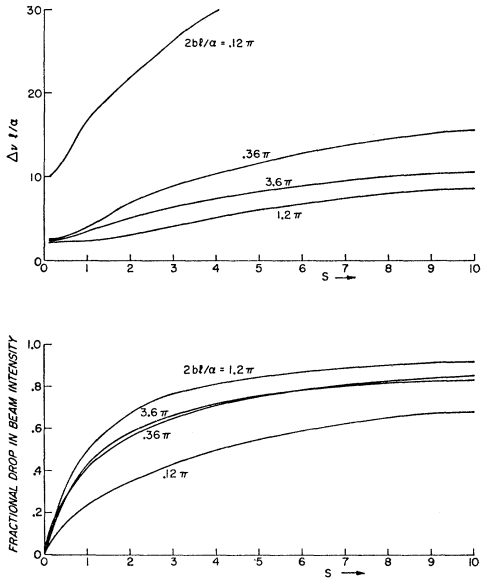

FIG. 4. Theoretical line width and intensity for single coil and slit. Optimal rotating field is given by $2bl/\alpha=1.2\pi$. The parameter S is a measure of the deflecting field strength.

$S=W_1$, so that this fraction is given by

$$f(\Delta m_J/J,J,V,S_1)=S_1J|\Delta m_J/J|/V^2 \quad \text{for} \quad S_1J|\Delta m_J/J|/V^2\leqslant 1$$
$$=1 \quad \text{for} \quad S_1J|\Delta m_J/J|/V^2\geqslant 1. \quad (10)$$

The signal, for a given V and J, is then given by

$$P(J,V,\theta,S_1)=\int_{-A}^{A}P\left(\frac{\Delta m_J}{J}\right)f\left(\frac{\Delta m_J}{J},J,V,S_1,\right)\frac{d\Delta m_J}{J}$$
$$=\int_0^A\left(\frac{1}{A}\right)f\left(\frac{\Delta m_J}{J},J,V,S_1\right)\frac{d\Delta m_J}{J} \quad (11)$$

where $A=2\sin\frac{1}{2}\epsilon$.

VI. VELOCITY AND J AVERAGING

The final signal observed must be obtained by averaging $P(J,V,\theta,S)$ over all the values of J and V, weighted according to their thermal distributions. If $J(J+1)$ is approximated by J^2 (as is consistent with the classical treatment) and the velocity dependence of the ionization probability for the electron bombarder is taken into account, these weighting factors are

$$W_J=\hbar^2/(2kTI)2J\exp(-\hbar^2/(2kTI)J^2)$$
$$=C^2 2J\exp(-C^2J^2) \quad (12)$$

$$W_V=(4/\sqrt{\pi})V^2\exp(-V^2) \quad (13)$$

so that

$$P(\theta)=\int_0^\infty\int_0^\infty\left(\frac{4}{\sqrt{\pi}}\right)V^2\exp(-V^2)C^2 2J$$
$$\times\exp(-C^2J^2)P(J,V,\theta,S_1)dJdV. \quad (14)$$

Since $P(J,V,\theta,S_1)$ has the form $f(JS_1A/V^2)$, the quantity C^2 may be incorporated into S_1 by setting $J'=CJ$, $S_1'=S_1/C$ to give

$$P(\theta)=\int_0^\infty\int_0^\infty\left(\frac{4}{\sqrt{\pi}}\right)V^2\exp(-V^2)2J'$$
$$\times\exp(-J'^2)P(J',V,\theta,S'_1)dJ'dV. \quad (15)$$

This double integral is impossible to evaluate analytically, even for simple cases, and cannot even be expressed easily in terms of a small number of calculable functions of single argument, as was done in the quantum mechanical spin-$\frac{1}{2}$ case.[6] It has hence been programmed for the IBM 7090 as a double Simpson's rule numerical integration.

The maximum intensity at resonance was found to be obtained when $2bl/\alpha=1.2\pi$, just as in the case of the quantum mechanical spin-$\frac{1}{2}$ case.[4] In terms of the rotating field H_1 this may be written

$$(l/\alpha)\omega_0H_1/H_0=1.2\pi. \quad (16)$$

The variation of linewidth and intensity with deflecting

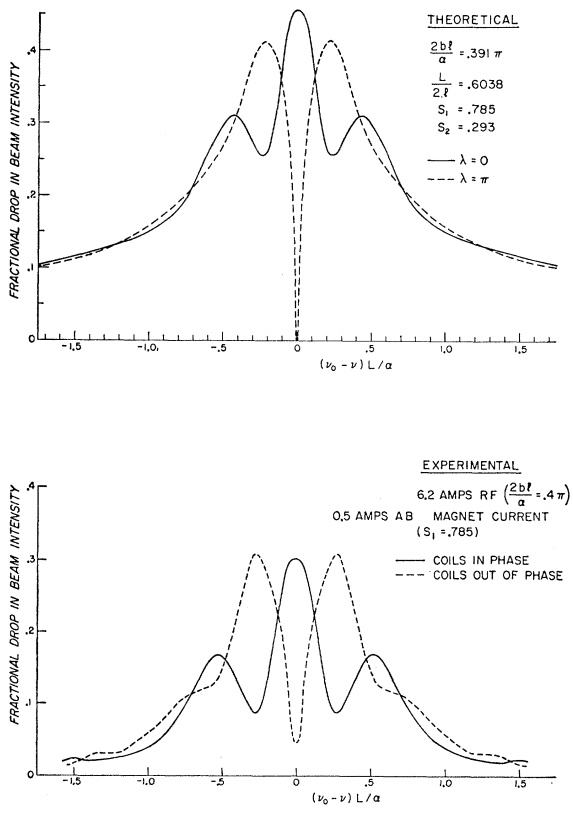

FIG. 5. Comparison between theoretical and experimental line shapes. Calculation is made for separated coils of length l each, separated by a gap L, with phase shift λ, and multiple slits.

[6] U. E. Kruse and N. F. Ramsey, J. Math. and Phys. **30**, 40 (1951).

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 nonadiabaticity 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

* This paper presents the results of one phase of research carried out at the Jet Propulsion Laboratory, California Institute of Technology under Contract NAS7-100, sponsored by the National Aeronautics and Space Administration.

[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).