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Atomic Oscillations in the Statistical Approximation*

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The dipolar spectral distribution function, or the photoabsorption cross section, of atoms is derived in the statistical approximation starting from general dynamic equations describing the density fluctuations induced in the atom by an external field. Using a local form of the theory it is found that coherence between the motion in different parts of the atom causes a modification of the spectral function. The modification can be put in the form of a dispersion denominator, and if this denominator becomes small in certain frequency ranges, the absorption is enhanced by what can be identified as collective resonances of the atom as a whole.

INTRODUCTION

THE results of an investigation of the quantum dynamics of atoms, reported in previous papers,[1] can be summarized briefly as follows. Atoms with more than one electron never are excited at the frequencies expected from a single-particle picture. The coupling between single-particle excitations always causes collective shifts to higher frequencies. The many-electron aspects of excitations from the outer and inner atomic shells, although characteristic and telling, do not change strongly the predictions of a single-particle picture. A one-to-one correspondence usually exists between the single-particle and the collectively shifted excitations from outer shells in the optical frequency range, which we shall call (1), of order 1 Ry, and those in the characteristic x-ray range (3), of order Z^2 Ry, Z being the atomic number; here conventional spectroscopic assignments are generally possible. By contrast, in an intermediate frequency range (2), of order Z Ry, the excitations from the intermediate atomic shells can be coupled so strongly that new collective resonances of the atom as a whole become possible.

A statistical approximation should apply best to the dynamics of atoms in just the frequency range (2) where atomic resonances can occur. Therefore, it is desirable to derive the dipolar spectral distribution function $g(\omega)$ for the statistical model, to complement the results obtained in this frequency range for the Hartree model. The photoabsorption cross section of atoms $\sigma(\omega)$ is directly proportional to $g(\omega)$,

$$\sigma(\omega)=[2\pi^2e^2/mc]g(\omega)\,,$$

where $[2\pi^2e^2/mc]=8.067\times10^{-18}$ Ry cm^2, if $\hbar$ Ry$=13.6$ eV. In the following we derive and discuss the spectral function $g(\omega)$ in the statistical approximation. A comparison with previous work in statistical approximations and with our results based on the Hartree approximation will be given elsewhere.

In Sec. 1, our basic integral equation is formulated in a self-consistent manner for the frequency-dependent density fluctuations in an atom set up by an external field of a definite frequency ω and of a wavelength which is long compared to atomic dimensions. The integral equation for dipolar oscillations is projected out in Sec. 2, since only this part need concern us in the present context. In Sec. 3, this equation is solved first in the limit of locally vanishing gradient of the electron density, to give us the atomic response in an essentially local approximation. Section 4 retains the dependence on density gradients. A dispersion denominator appears which accounts for the absorption by oscillations of the atom as a whole.

1. Integral Equation for Atomic Oscillations in the Statistical Approximation

We consider the dynamics of an atom in the presence of an external field of wavelength long compared with atomic dimensions. The field induces fluctuations in the electron density about the unperturbed density $\rho_0(\mathbf{x})$. We assume that each volume element responds to the total field acting on it as if it were part of an infinite electron gas of the same density.

First we consider an electron gas enclosed in a box of volume Ω. The total field is denoted by $V(\mathbf{x},\omega)$ and the induced electron density by $\rho(\mathbf{x},\omega)$. The corresponding Fourier amplitudes, defined with respect to the box, are

$$\rho_\Omega(\mathbf{k},\omega)=\int_\Omega d^3x e^{i\mathbf{k}\cdot\mathbf{x}}\rho(\mathbf{x},\omega) \tag{1}$$

and

$$V_\Omega(\mathbf{k},\omega)=\int_\Omega d^3x e^{i\mathbf{k}\cdot\mathbf{x}}V(\mathbf{x},\omega)\,, \tag{2}$$

respectively.

We postulate a linear relation between the induced

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[1] W. Brandt and S. Lundqvist, Phys. Letters 4, 47 (1963); Phys. Rev. 132, 2135 (1963); Arkiv Fysik 28, 399 (1965); henceforth referred to as I, II, and III, respectively.

electron density and the total field of the form

$$\rho_\Omega(\mathbf{k},\omega)=H(\mathbf{k},\omega,\rho_0)V_\Omega(\mathbf{k},\omega). \tag{3}$$

The dispersive properties of the medium with respect to frequency and wave number (or time and space) are embodied in the function $H(\mathbf{k},\omega,\rho_0)$ which, for the present, we shall assume to be known.

We now divide the atom into a number of volume elements Ω_i, each of which is small enough so that the density is constant, but large enough so that it contains many electrons. The total field at a point $\mathbf{x}$ is the sum of the external field $V_{\text{ext}}(\mathbf{x},\omega)$ and the field due to the induced electron density, which we write as

$$V_{\text{ind}}(\mathbf{x},\omega)=\frac{1}{(2\pi)^3}\int d^3q e^{-i\mathbf{q}\cdot\mathbf{x}}\frac{4\pi e}{q^2}\rho(\mathbf{q},\omega), \tag{4}$$

where $\rho(\mathbf{q},\omega)$ is the Fourier component of the total induced electron density,

$$\rho(\mathbf{q},\omega)=\sum_i \rho_{\Omega_i}(\mathbf{q},\omega). \tag{5}$$

Because of Eqs. (2) and (3), the induced density in Ω_i is given by

$$\rho_{\Omega_i}(\mathbf{k},\omega)=H(\mathbf{k},\omega,\rho_0)\int_{\Omega_i} d^3x e^{i\mathbf{k}\cdot\mathbf{x}} \times[V_{\text{ind}}(\mathbf{x},\omega)+V_{\text{ext}}(\mathbf{x},\omega)]. \tag{6}$$

Summing over all volume elements and formally making the transition to infinitesimals we obtain the following integral equation:

$$\begin{aligned}\rho(\mathbf{k},\omega)=(2\pi)^{-3}\int d^3x\int d^3q H(\mathbf{k},\omega,\rho_0(\mathbf{x}))\\ \times e^{i(\mathbf{k}-\mathbf{q})\cdot\mathbf{x}}[4\pi e/q^2]\rho(\mathbf{q},\omega)\\ +\int d^3x H(\mathbf{k},\omega,\rho_0(\mathbf{x}))e^{i\mathbf{k}\cdot\mathbf{x}}V_{\text{ext}}(\mathbf{x},\omega).\end{aligned} \tag{7}$$

In the following we work exclusively with the space representation of this equation, which we write in the form

$$\rho(\mathbf{x},\omega)=\int d^3x' K(\mathbf{x},\mathbf{x}',\omega)\rho(\mathbf{x}',\omega) +\int d^3x' H(\mathbf{x},\mathbf{x}',\omega)V_{\text{ext}}(\mathbf{x}',\omega), \tag{8}$$

where

$$K(\mathbf{x},\mathbf{x}',\omega)=\frac{1}{(2\pi)^3}\int d^3q\frac{4\pi e}{q^2}H(\mathbf{q},\omega,\rho_0(\mathbf{x}))e^{-i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}, \tag{9}$$

and

$$H(\mathbf{x},\mathbf{x}',\omega)=\frac{1}{(2\pi)^3}\int d^3q H(\mathbf{q},\omega,\rho_0(\mathbf{x}))e^{-i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}. \tag{10}$$

We note that the form of Eq. (8) is perfectly general within the linear-response approximation and that the electron-gas approximation appears explicitly only in Eqs. (9) and (10). As written, the kernel $K(\mathbf{x},\mathbf{x}',\omega)$ is not symmetric. However, it is inherent in the statistical approximation that the unperturbed density does not change appreciably over the distance of nonlocality in space. Therefore, we are justified in considering the kernel to be symmetric in the following.

2. Radial Integral Equation for Dipolar Oscillations

Take an atom with spherical symmetry in the ground state, i.e., $\rho_0(\mathbf{x})=\rho_0(r)$. We apply a slowly varying external field of the form $E=E_0\exp(-i\boldsymbol{\kappa}_0\cdot\mathbf{x})$, so that

$$V_{\text{ext}}(\mathbf{x},\omega)=(i\kappa_0)^{-1}eE_0e^{-i\kappa_0\cdot\mathbf{x}}. \tag{11}$$

Using Eqs. (8) and (10) we obtain

$$p(\mathbf{x},\omega)=\int d^3x' e^{i\kappa_0\cdot(\mathbf{x}-\mathbf{x}')}K(\mathbf{x},\mathbf{x}',\omega)p(\mathbf{x}',\omega) -\kappa_0^{-2}eE_0H(\boldsymbol{\kappa}_0,\omega,\rho_0(\mathbf{x})) \tag{12}$$

with

$$p(\mathbf{x},\omega)=(i\kappa_0)^{-1}\rho(\mathbf{x},\omega)e^{i\kappa_0\cdot\mathbf{x}}. \tag{13}$$

We now consider the limit of long wavelengths such that $\kappa_0 a\ll 1$, where a is the linear extension of the atom. In this limit Eq. (12) takes the simpler form

$$p(\mathbf{x},\omega)=\int d^3x' K(\mathbf{x},\mathbf{x}',\omega)p(\mathbf{x}',\omega)-\frac{E_0e\rho_0(\mathbf{x})}{\omega^2}. \tag{14}$$

Considering density oscillations of dipolar symmetry and long wavelength we can consider solutions of the form

$$p(\mathbf{x},\omega)=[f(r)/r]\cos^2\theta, \tag{15}$$

where θ is the angle relative to the direction of the external field. Observing that the inhomogeneous term in Eq. (14) has spherical symmetry, we obtain the following radial integral equation for the forced vibration of the atom:

$$f(r)=\int_0^\infty dr' K(r,r')f(r')-\frac{3E_0e\rho_0(r)r}{\omega^2}. \tag{16}$$

The kernel in Eq. (16) is given by the formula

$$K(r,r')=\int_0^\infty dq\frac{4\pi e}{q^2}H(q,\omega,r)\varphi(qr)\varphi(qr'), \tag{17}$$

where

$$\varphi(qr)=(2/\pi)^{1/2}\sin qr. \tag{18}$$

From the solution of Eq. (16) we can calculate the induced dipole moment P using Eqs. (13) and (15), from which the frequency-dependent polarizability is obtained as the coefficient of the external field E_0. Thus,

$$P(\omega)=e\int p(\mathbf{x},\omega)d^3x=\frac{4\pi e}{3}\int_0^\infty f(r)r dr=\alpha(\omega)E_0. \tag{19}$$

3. The Solution of the Integral Equation in the Zero-Gradient Approximation

We consider the equation

$$f(r)=f_0(r)+\int_0^\infty dr'\,K(r,r',\omega)f(r'), \tag{20}$$

with

$$K(r,r',\omega)=\int_0^\infty dq\frac{4\pi e}{q^2}H(q,\omega,\rho_0(r))\varphi(qr)\varphi(qr') \tag{21}$$

and

$$f_0(r)=-[3E_0e\rho_0(r)r/\omega^2], \tag{22}$$

$\varphi(qr)$ being defined by Eq. (18).

We now introduce the notation

$$\chi(q^2,\omega,\rho_0(r))=[4\pi e/q^2]H(q,\omega,\rho_0(r)) \tag{23}$$

and using the relation

$$\int dq\,q^{2n}\varphi(qr)\varphi(qr')=(-1)^n\delta^{(2n)}(r-r') \tag{24}$$

we can write the kernel symbolically as

$$\begin{aligned}K(r,r',\omega)&=\int_0^\infty dq\,\chi(q^2,\omega,\rho_0(r))\varphi(qr)\varphi(qr')\\&=\chi[-d^2/dr^2,\omega,\rho_0(r)]\delta(r-r')\end{aligned} \tag{25}$$

with the understanding that we shall differentiate only with regard to the variable r in the δ-function, and not with regard to r in the unperturbed density $\rho_0(r)$. This implies neglecting the gradient of the unperturbed density. We call this the *zero-gradient approximation.* In this approximation the integral equation is solved by first writing the symbolic equation

$$f(r)=f_0(r)+\chi(-d^2/dr^2,\,\omega,\,\rho_0(r))f(r) \tag{26}$$

with the formal solution

$$f(r)=f_0(r)/[1-\chi(-d^2/dr^2,\,\omega,\,\rho_0(r))] \tag{27}$$

which, by again using Eq. (24), can be written as

$$\begin{aligned}f(r)&=\int_0^\infty dq\int_0^\infty dr'\frac{\varphi(qr)\varphi(qr')}{1-\chi(q^2,\omega,\rho_0(r))}f_0(r')\\&=\int_0^\infty dq\int_0^\infty dr'\frac{\varphi(qr)\varphi(qr')}{\epsilon(q,\omega,\rho_0(r))}f_0(r'),\end{aligned} \tag{28}$$

where we have introduced explicitly the wave vector and frequency dependent dielectric function

$$\epsilon(q,\omega,\rho_0(r))=1-\chi(q^2,\omega,\rho_0(r)) \tag{29}$$

of a uniform gas of density $\rho_0(r)$.

Because of Eqs. (19), (22), and (28) we obtain the dipole moment

$$P(\omega)=-\frac{4\pi e^2E_0}{\omega^2}\int r dr\,r'dr'\,dq\frac{\varphi(qr)\varphi(qr')}{\epsilon(q,\omega,\rho_0(r))}\rho_0(r'), \tag{30}$$

and derive the polarizability

$$\alpha(\omega)=-4\pi e^2\int_0^\infty r dr\int_0^\infty dq\frac{\varphi(qr)}{\omega^2\epsilon(q,\omega,\rho_0(r))}\times\int_0^\infty r'dr'\,\varphi(qr')\rho_0(r'). \tag{31}$$

This expression shows the characteristic feature of this approximation. The absorption in a given radial shell will be determined by the imaginary part of the inverse of the dielectric function, and the absorption will at each frequency generally have its maximum around some wave number q_0. If we neglect the imaginary part of $\epsilon(q,\omega,\rho_0(r))$ the absorption will take place at a definite momentum q_0 (assumed to be real), given by the solution of

$$\epsilon(q_0,\omega,\rho_0(r))=0. \tag{32}$$

Thus, the resonance condition is locally the same as that for a plasma resonance in a uniform electron gas. This feature is a direct consequence of neglecting the explicit effect of a density gradient.

Taking the imaginary part of the integral we obtain for the oscillator-strength distribution function

$$g(\omega)=\frac{2m}{\pi e^2}\omega\,\mathrm{Im}\alpha(\omega)=\frac{8\pi}{\omega}\int r dr\frac{\varphi(q_0r)}{|d\chi/dq|_{q_0}}\times\int r'dr'\,\varphi(q_0r')\rho_0(r'), \tag{33}$$

where q_0 has to be determined from Eq. (32), and the integration extends over the region, where $q_0=q_0(\omega,r)$ is real.

We shall in the following discuss a few simple approximations for $\epsilon(q,\omega,\rho_0)$ deferring the treatment in terms of the full expressions given by Lindhard[2] to a later paper.

The simplest approximation is to neglect the spatial dispersion and describe the local response by the Drude approximation

$$\chi(q^2,\omega,\rho_0(r))=\omega_0^2(r)/\omega^2, \tag{34}$$

where $\omega_0(r)=[4\pi e^2\rho_0(r)/m]^{1/2}$ is the local plasma frequency. For the polarizability we obtain

$$\alpha(\omega)=\frac{e^2}{m}\int d^3x\frac{\rho_0(\mathbf{x})}{\omega_0^2(\mathbf{x})-\omega^2} \tag{35}$$

and the oscillator strength distribution is given by

$$g(\omega)=\int d^3x\,\rho_0(\mathbf{x})\delta(\omega_0(\mathbf{x})-\omega). \tag{36}$$

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[2] J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 28, No. 8 (1954).

As pointed out in II, the effect of the Drude screening is to replace the free-particle response by one where each part of the medium behaves as if it had a characteristic frequency equal to the local plasma frequency. The absorption at a given frequency occurs in an infinitesimally thin shell, determined by the condition $\omega=\omega_0(r)$. Resonances or detailed structure in the absorption spectrum will only occur where the density varies slowly.

Equation (36) is of interest as a reference function for studying changes in $g(\omega)$ due to atomic oscillations. We have evaluated Eq. (36) for different density distributions $\rho_0(r)$. The resulting photoabsorption cross sections are shown in Fig. 1 on the reduced frequency scale ω/Z Ry for the Thomas-Fermi and the Lenz-Jensen density distributions of the statistical atom,[3] and for the Hartree density distributions of Ar and Hg. Clearly, as pointed out in II, this local approximation essentially maps the corresponding charge distributions. The shell structure of the Hartree atoms is reflected in modulations of the photoabsorption cross sections relative to the curves for the statistical atoms which give a good average of the frequency range (2) of interest, i.e., for $\omega/Z\ \text{Ry}>0.01$. The slow decline of the density in the Thomas-Fermi atom as $r\rightarrow\infty$ causes the spectral function to rise to the constant value

$$g(0)=(3/2)^{3/2}\pi\ \text{Ry}^{-1},$$

corresponding to $\sigma(0)=46.56\times10^{-18}\ \text{cm}^2$. The curves for the Lenz-Jensen atom and the Hartree atoms pass through a maximum near the lower end of our range (2) and go to zero with the frequency. At very high frequencies the spectral functions of the Hartree atoms extend only to a cutoff frequency corresponding to the electron density at the origin. The local spectral functions of the Thomas-Fermi and the Lenz-Jensen atoms nearly coelesce and decline asymptotically as $(\omega/Z\ \text{Ry})^{-3}$. By coincidence the spectral functions of real atoms at very high frequencies also fall off approximately as ω^{-3}.

We next consider the case of slight dispersion which follows from the long-wavelength plasma motion.

$$\chi(q,\omega,r)=\omega_0^2(r)/\omega^2+\tfrac{3}{5}v_0^2(r)q^2/\omega^2; \tag{37}$$

$v_0(r)$ denotes the Fermi velocity for the electrons at r. A formula which only differs slightly by a numerical coefficient (5/9 instead of 3/5) follows from the dynamic Thomas-Fermi theory as developed by Bloch.[4] The solution of Eq. (32) gives for the local wave number

$$q_0^2=\frac{\omega^2-\omega_0^2(r)}{\tfrac{3}{5}v_0^2(r)}. \tag{38}$$

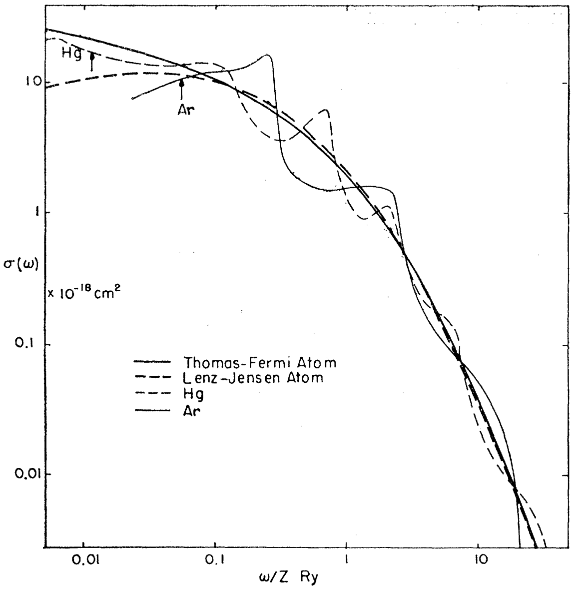

FIG. 1. Photoabsorption cross sections as a function of ω/Z Ry, calculated in the extreme local approximation, Eq. (36), for the electron-density distributions of the statistical Thomas-Fermi and Lenz-Jensen atoms and for the Hartree distributions of Ar and Hg.

Inserting this expression in Eq. (33) yields

$$g(\omega)=4\pi\omega\int rdr\frac{|q_0|\,\varphi(q_0r)}{\omega^2-\omega_0^2(r)}\int r'dr'\ \varphi(q_0r')\rho_0(r'), \tag{39}$$

where the integration extends over the regions, where q_0 is real.

At a given frequency ω the radial integrations extend from a radius r_0 corresponding to $q_0=0$, viz., where $\omega=\omega_0(r_0)$. For $r>r_0$ the local wave number q_0 increases without limit as $r\rightarrow\infty$. However, the contribution from the low-density region to $g(\omega)$ is small because the unperturbed density tends to zero exponentially. This approximation has the same qualitative characteristics as the theory of Wheeler and Fireman[5] and Ball.[6]

We finally give the result for a simple interpolation formula given by Lindhard to represent approximately the semiclassical domain over a larger range than Eq. (37).

$$\chi(q,\omega,\rho_0(r))=\frac{\omega_0^2(r)}{\omega^2-\tfrac{3}{5}v_0^2(r)q^2}. \tag{40}$$

From Eq. (32) we again obtain for the local wave number

$$q_0^2(r)=\frac{\omega^2-\omega_0^2(r)}{\tfrac{3}{5}v_0^2(r)}, \tag{41}$$

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[3] See, e.g., P. Gombas, *Die Statistische Theorie des Atoms und ihre Anwendungen* (Springer-Verlag, Berlin, 1949).

[4] F. Bloch, Z. Physik **81**, 363 (1933).

[5] J. A. Wheeler and E. L. Fireman, Aeronutronic System Publication U-099, 1957 (unpublished).

[6] J. A. Ball, thesis, Princeton, University, 1963 (unpublished).

but now $g(\omega)$ becomes

$$g(\omega)=4\pi\omega\int rdr\frac{\omega_0{}^2(r)}{\omega^2}\frac{|q_0|\,\varphi(q_0r)}{\omega^2-\omega_0{}^2(r)}\times\int r'dr'\;\varphi(q_0r')\rho_0(r'),\qquad(42)$$

where, as in the previous case, the integration extends over the interval $r>r_0$, with r_0 the radius where $q_0(r)=0$. Here the suppression of the short-wavelength contributions from the low-density region is considerably stronger than in Eq. (39).

We note that these three approximate formulas, Eqs. (36), (39), and (42) represent local approximations in the sense that the resonance condition (32) applies locally. At the same time, however, the description of the response is purely collective, since the resonance condition implies the vanishing of the dielectric function, which is taken as that for an extended uniform medium of the corresponding density. Because of spatial dispersion, as reflected in the q dependence of $\chi(q,\omega,\rho_0(r))$, there is a characteristic nonlocality length involved. In order for our approximation to be meaningful, this nonlocality length must be small compared to the distance over which the density changes appreciably.

According to the scheme discussed in this section, the absorption is a continuous function of frequency; at each point absorption will take place at a given frequency ω provided the resonance condition (32) is fulfilled for a real wave number q_0. One effect of the density gradient, which was neglected in this section, will be to dynamically couple different parts of the atom in such a way that the polarizability and spectral distribution can no longer be obtained by adding the contributions from the different spherical shells. This coupling may not only give a modulation of the type of results just discussed, it may also bring in the effects of coherent motion in the atom, which would manifest itself in the occurrence of new resonances. A modification to include such effects is discussed in the following section.

4. Solution of the Integral Equation in the Small-Gradient Approximation

We rewrite the integral equation (20) by introducing formally a strength parameter λ.

$$f(r)=f_0(r)+\lambda\int dr'\;K(r,r')f(r').\qquad(43)$$

The Fredholm solution is

$$f(r)=f_0(r)+\int dr'\frac{D(r,r',\lambda)f_0(r')}{D(\lambda)},\qquad(44)$$

where $D(r,r',\lambda)$ and $D(\lambda)$ denote Fredholm's first minor and determinant, respectively. The solution to our problem is obtained by letting $\lambda\rightarrow1$.

The approximation discussed in the preceding section corresponds to setting $D(\lambda)=1$ and

$$D(r,r',\lambda)=\int\frac{\lambda\chi(q,\omega,r)}{1-\lambda\chi(q,\omega,r)}\varphi(qr)\varphi(qr')dq.\qquad(45)$$

The solution corresponding to $D(\lambda)=1$ and Eqs. (44) and (45) is in general exact only in the limiting cases of zero spatial dispersion or zero density gradient. For zero spatial dispersion the kernel is diagonal and the solution is trivial [cf., Eq. (28)]. Taking spatial dispersion into account, we have $D(\lambda)\neq1$ in a medium with a density gradient. In addition we get correction terms to Eq. (45). Still as long as the density gradient is considered to be small one can assume Eq. (45), and calculate $D(\lambda)$ from the Fredholm relation[7]

$$-\lambda D'(\lambda)=\int dr\;D(r,r,\lambda).\qquad(46)$$

The calculation is elementary and gives the result

$$D(1)=1+F(\omega),$$

where

$$F(\omega)=\int dr\int dq\;\Phi(qr)\frac{1}{1-\chi(q,\omega,r)}\frac{d\chi(q,\omega,r)}{dr},\qquad(47)$$

with

$$\Phi(qr)=\int_0^r|\varphi(qr')|^2dr'.\qquad(48)$$

The solution in the small-gradient approximation therefore is given by the formula

$$f(r)=\int dqdr'\frac{\varphi(qr)\varphi(qr')}{1-\chi(q,\omega,r)}f_0(r')\Big/1+F(\omega).\qquad(49)$$

From Eq. (49) the function $g(\omega)$ can be found.

The difference between Eqs. (49) and (28) lies in the frequency-dependent modulation given by the denominator. If, in certain frequency ranges, the absolute magnitude of the denominator becomes small the absorption is enhanced because of what may be interpreted as a collective oscillation in the atom. If the denominator of Eq. (49) in fact were to vanish at some frequency, the homogeneous equation to Eq. (43) has a solution corresponding to a density oscillation. In this sense a small denominator indicates that such an oscillation represents an approximate eigenstate of the atom.

A similar formula was derived by the authors in II. The formula given here differs in two respects. In the first place, in II we did not separate the equations into

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[7] W. V. Lovitt, *Integral Equations* (Dover Publications, Inc., New York, 1924).

angular-momentum components which we have done here, and secondly the derivation of the small-gradient formula given here is more straightforward.

If we had calculated $D(1)$ using the first order approximation to the Fredholm determinant

$$D(1)\approx 1-\int dr\, K(r,r)\,, \tag{50}$$

we would have obtained

$$D(1)\approx 1+\int dr \int dq\, \Phi(qr)\frac{d\chi(q,\omega,r)}{dr}\,. \tag{51}$$

Comparing this with Eq. (47) we see that the screening, expressed by the local dielectric constant $\epsilon(q,\omega,r)$, is included in Eq. (47) but is neglected in Eq. (51). That is, the formula Eq. (47) amounts to a summation of a class of terms in the Fredholm expansion of $D(\lambda)$, which corresponds to the summation of ring diagrams in the electron-gas problem but neglects, e.g., effects due to higher derivatives and powers of the density gradient. We note that

$$d\chi(q,\omega,r)/dr \rightarrow 0$$

with the density gradient, i.e., in the limit of zero-gradient density the correct result for a uniform electron gas is guaranteed.

We have begun comprehensive calculations of the statistical spectral functions derived in this paper.

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Mode Competition and Frequency Splitting in Magnetic-Field-Tuned Optical Masers*

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Equations are given describing the beat-frequency variation and mode competition in a gaseous optical maser operated in a magnetic field parallel to the maser axis. The equations include only lowest order nonlinear terms. Important terms in the amplitude- and frequency-determining equations are shown to arise from an induced atomic precession. These terms have a character similar to those describing the effects of selective depletion of the velocity distribution or "hole burning." It is shown that the induced atomic precession causes parametric conversion of an optical field of one circular polarization into one of the other polarization with a frequency shift equal to the rate of precession. This process tends to make the competition between modes of different polarizations important. An additional feature, not found in the scalar theory, is that, for sufficiently large magnetic fields, competition can be important between modes separated in frequency by several Doppler-broadened linewidths.

THIS paper gives equations describing the beat-frequency variation and mode competition in a gaseous optical maser operated in a magnetic field parallel to the maser axis, the equations being obtained from a density-matrix analysis similar to that of W. E. Lamb, Jr.[1,2] Important terms in the amplitude- and frequency-determining equations arise from an induced atomic precession in atoms occupying regions of the velocity distribution analogous to the population depletion holes described by Bennett, Jr.,[3] and by Lamb.[1] Similar calculations by Tang and Statz,[4] and by Culshaw and Kannelaud[5] do not produce the explicit equations given here.

Assuming a $J=1$ to $J=0$ optical transition, circularly polarized components of the optical field of amplitude E_+ and E_-, only one cavity resonance (Ω_+,Ω_-) above oscillation threshold for each polarization (Ω_+ is not necessarily the same resonance as Ω_-), an active medium filling the entire cavity, and otherwise following the assumptions and method of Lamb, one obtains the amplitude- and frequency-determining equations

$$\alpha_+-\beta_+E_+{}^2-\theta_{+-}E_-{}^2=0\,, \tag{1a}$$

$$\alpha_--\beta_-E_-{}^2-\theta_{-+}E_+{}^2=0\,, \tag{1b}$$

$$\nu_+=\Omega_++\sigma_++\rho_+E_+{}^2+\tau_{+-}E_-{}^2\,, \tag{2a}$$

$$\nu_-=\Omega_-+\sigma_-+\rho_-E_-{}^2+\tau_{-+}E_+{}^2\,, \tag{2b}$$

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[*] The work of one of the authors (M.S.) was supported by the U. S. Air Force Office of Scientific Research.

[1] W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).

[2] A more complete theory allowing x-y asymmetries in the resonator, transverse magnetic fields, and larger J values is being carried out by the authors in association with W. E. Lamb, Jr.

[3] W. R. Bennett, Jr., Appl. Opt., Suppl. **1**, 24 (1962).

[4] C. L. Tang and H. Statz, Phys. Rev. **128**, 1013 (1962).

[5] W. Culshaw and J. Kannelaud, Phys. Rev. **136**, A1209 (1964).