Higher-order nonlinear susceptibilities for generation of optical radiation harmonics in atomic gases

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A general investigation is reported of nonlinear susceptibilities of atoms governing nonresonant generation of higher optical harmonics. An analysis is made of higher-order (in respect of the field) corrections to the susceptibility calculated in the first nonvanishing order of perturbation theory and it is shown that these corrections can be observed experimentally. The results are given of a numerical calculation of the tensors $\chi^{(5)}_{ijklm_n}(-5\omega;\omega,\omega,\omega,\omega,\omega)$ and $\chi^{(5)}_{ijklm_n}(-3\omega;-\omega,\omega,\omega,\omega,\omega)$ for hydrogen, alkali atoms, and rare gases at typical laser frequencies. A comparison is made with the available experimental data.

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§1. INTRODUCTION

In the decade from the first experimental observation of the generation of the third harmonic of optical radiation in atomic gases[1] the progress in experimental research has been so fast that it is now possible to observe nonlinear optical effects in atoms due to nonlinear susceptibilities of orders 9–11 (Ref. 2). This progress has been stimulated by the fact that atomic gases can be used to convert the frequencies of high-power coherent radiation from the optical to the ultraviolet and soft x-ray ranges. The use of atomic gases for these purposes has a number of advantages over the use of other nonlinear media (crystals, liquids) and, in particular, the generation of harmonics with the aid of atoms has made it possible to obtain coherent radiation of record energy amounting to $\omega \sim 33$ eV (Ref. 3).

In contrast to the lowest (third-order) nonlinearities, on which information is available for a large number of atoms at different frequencies (see, for example, Refs. 4 and 5), higher-order susceptibilities corresponding to the generation of the fifth and higher harmonics have been investigated much less thoroughly (see Akhmanov's review[2]). There are only approximate calculations[6,7] of these quantities and these have been carried out on the basis of a fairly rough approximation in which only finite number of terms is retained in the sums over intermediate states in the matrix elements of perturbation theory. Moreover, no analyses have yet been made of the contribution of the higher-order (in respect of the field) effects to the susceptibility calculated in the first nonvanishing order of perturbation theory. This is of practical importance in harmonic generation because intensities of pump fields used in experiments on alkali atoms and rare gases reach $\sim 10^{12}-10^{15}$ W/cm^2 in the picosecond regime. In such strong fields we can expect not only manifestation of higher-order effects in perturbation theory but also have to consider whether it is permissible to describe harmonic generation by a scheme traditional in nonlinear optics and based on the expansion of the polarization vector of the medium as a series in powers of the external field.

We shall use the method of a model potential, developed earlier[5] for many-photon processes in atoms, to analyze off-resonance susceptibilities of atoms describing the scattering of higher optical radiation harmonics (§2) and also to consider nonlinear corrections to the susceptibilities in a strong pump field (§3). Numerical calculations will show that these corrections can be observed experimentally at pump intensities which are being currently used and, in particular, that such corrections can give rise to a considerable difference between the polarization of the generated radiation and the pump polarization in the case of elliptic polarization of the latter. Specific calculations will be made for the hydrogen atom, and also for alkali atoms and rare gases, which are the main elements used in experimental generation of harmonics in atomic gases.

§2. SCATTERING AT A FREQUENCY $n\omega$ IN THE FIRST NONVANISHING ORDER IN RESPECT OF THE FIELD

The pump field **F** will be regarded as monochromatic and elliptically polarized in the xy plane so that

$$\mathbf{F}(r,t)=F\mathrm{Re}\{\mathbf{e}e^{-i\omega t+ikz}\},\ \mathbf{e}=\frac{\mathbf{e}_x-i\xi\mathbf{e}_y}{(1+\xi^2)^{1/2}},\ -1\leqslant\xi\leqslant 1. \tag{1}$$

The degrees of linear (l) and circular (A) polarizations of the field are

$$l=\frac{1-\xi^2}{1+\xi^2},\ A=\frac{2\xi}{1+\xi^2}.$$

The wave intensity $I_\omega = cF^2/8\pi$ is independent of ξ.

The dipole moment of an atom induced by the field **F**

$$\mathbf{D}(t)=\mathrm{Re}\left\{\sum_n \mathbf{P}(n\omega)e^{-in\omega t}\right\} \tag{2}$$

considered in the electric dipole approximation contains only the odd harmonics ($n=1,3,\ldots$). The vector $\mathbf{P}(n\omega)$ can be calculated only within the framework of perturbation theory of the interaction of an atom with the field and it reduces to a calculation of the nonlinear susceptibility tensors, which are coefficients in the expansion of $\mathbf{P}(n\omega)$ in powers of F. It is usual to retain only the first nonvanishing order in respect of F, when **P** is of the form (see, for example, Ref. 8)

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$$P_i(n\omega) \equiv P_i^{(n)}(n\omega) = \frac{F^n}{2^{n-1}} \sum_{j_1 \dots j_n} \chi^{(n)}_{ij_1 \dots j_n}(-n\omega; \omega, \dots, \omega) e_{j_1} \dots e_{j_n}. \quad (3)$$

Here,

$$\chi^{(n)}_{ij_1 \dots j_n}(-n\omega; \omega, \dots, \omega) = T(n\omega, (n-1)\omega, (n-2)\omega, \dots, \omega) + T(-\omega, (n-1)\omega, (n-2)\omega, \dots, \omega) + \dots + T(-\omega, -2\omega, \dots, -n\omega) \quad (4)$$

is the component of the nonlinear susceptibility tensor containing $(n+1)$-term matrix element T of order n given by

$$T(\omega_1, \dots, \omega_n) = \langle 0 | \hat{d}_i G_{E_0+\omega_1} \hat{d}_{j_1} G_{E_0+\omega_1+\omega_2} \dots G_{E_0+\omega_n} \hat{d}_{j_n} | 0 \rangle, \quad (5)$$

where $\hat{d}$ and G_E are the dipole moment operator and the Green function of the atom, and E_0 is the energy of the initial state $|0\rangle$.

For simplicity, we shall confine ourselves to the most important case when the scattering atom is in a nondegenerate ground state $\psi_0(\mathbf{r}) \equiv |0\rangle$. In this case the tensor χ has only one linearly independent component

$$\chi^{(n)}_{xx \dots x}(-n\omega; \omega, \dots, \omega) = \chi^{(n)}(-n\omega),$$

whereas the other components can be expressed in terms of $\chi^{(n)}$ using the symmetry relationships. In particular,

$$\chi^{(n)}_{yyx \dots x}(-n\omega; \omega, \dots, \omega) = \frac{1}{n} \chi^{(n)}(-n\omega),$$

$$\chi^{(n)}_{yyzz \dots x}(-n\omega; \omega, \dots, \omega) = \frac{1}{n(n-2)} \chi^{(n)}(-n\omega).$$

Any transposition of the indices does not alter the value of the corresponding component, and the components with an odd number of any of the indices x, y, and z vanish.

These relationships allow us to rewrite Eq. (3) in the vector form

$$\mathbf{P}^{(n)}(n\omega) = \mathbf{e} F \left(\frac{l F^2}{4} \right)^{(n-1)/2} \chi^{(n)}(-n\omega). \quad (6)$$

Thus, the polarization $\mathbf{P}^{(n)}(n\omega)$ is identical with the polarization e of the pump wave. In the case of circular polarization ($l = 0$) the polarization vector is $\mathbf{P}(n\omega) = 0$ for $n > 1$ and there is no scattering at the harmonic frequency, which is due to the specific nature of the selection rules for a circularly polarized field.

The angular distribution of the radiation emitted by an atom at a frequency $\omega' = n\omega$ is governed by the scattering cross section in the direction $\mathbf{n}'$:

$$\frac{d\sigma(n\omega)}{d\Omega_{n'}} = \frac{1}{I_0} \frac{(n\omega)^4}{8\pi c^2} |[\mathbf{P}^{(n)}(n\omega)\mathbf{n}']|^2 = \left(\frac{l F^2}{4} \right)^{n-1} \left(\frac{n\omega}{c} \right)^4 |\chi^{(n)}(-n\omega)|^2 (1 - |\mathbf{e}\mathbf{n}'|^2). \quad (7)$$

[Substitution of $n = 1$ in Eq. (7) gives the Rayleigh scattering cross section; $\chi^{(1)}(-\omega)$ is the dynamic polarizability of an atom.] The cross section $d\sigma/d\Omega_n$ has its maximum value in the direction of propagation of the incident radiation, so that we can make effective use of the scattering process at a frequency $n\omega$ for coherent generation of the n-th harmonic in atomic gases ensuring coherence by matching the phases of the incident and generated radiations through introduction of a buffer gas.[9]

The generated harmonic radiation reaches a considerable intensity and can itself induce nonlinear polarization in the investigated medium and give rise to cascade-like generation of higher harmonics as a result of nonlinear susceptibilities of lower orders.[2,10] In atomic gases this situation is possible beginning from $n = 5$. However, the influence of cascade processes is significant only when the atomic medium is not only phase-matched for the fundamental-frequency radiation and the n-th harmonic, but also for at least one of intermediate frequencies $(n-2)\omega, \dots, 3\omega$. In practice this condition can only be satisfied accidentally so that cascade processes in atomic gases cannot usually be coherent simultaneously with direct processes, so that we shall ignore the corresponding inhomogeneities.

TABLE I. Nonlinear susceptibility tensors of atoms at typical laser frequencies ($\omega_N = 9440$ cm^{-1} = 0.043 a.u., $\omega_R = 14\,400$ cm^{-1} = (0.0656 a. u.). The numbers in parentheses give powers of ten (10^n). Conversion relationships: $(\chi^{(3)})$ a.u. = 1.985×10^{39} $(\chi^{(3)})$ cgs, $(\chi^{(5)})$ a.u. = 5.84×10^{53} $(\chi^{(5)})$ cgs.

Atom	ω	$\chi^{(3)}(-3\omega)$	$\chi^{(5)}(-5\omega)$	$\chi^{(5)}_{\parallel}(-3\omega)$	$\chi^{(5)}_{\perp}(-3\omega)$
H	ω_N	278.6	9.14(4)	4.90(4)	1.135(4)
	ω_R	391.4	8.67(5)	1.11(5)	3.40(4)
	$2\omega_N$	658.8	−1.24(7)	5.68(5)	3.19(5)
	$2\omega_R$	−8784	−6.10(5)	−5.50(6)	1.05(6)
He	ω_N	9.66	205	183	36.8
	ω_R	10.3	290	215	45.4
	$2\omega_N$	11.2	465	262	60.2
	$2\omega_R$	15.1	3.53(3)	572	175
	$4\omega_N$	23.6	−1.98(4)	2.36(3)	1.14(3)
Li	ω_N	−1.11(5)	4.8(9)	−3.9(9)	−6.8(9)
	ω_R	3.67(6)	−1.0(9)	9.1(12)	−1.6(13)
Na	ω_N	−1.79(6)	1.0(10)	3.1(10)	−1.3(10)
	ω_R	1.83(6)	1.5(10)	−1.2(11)	−3.9(10)
K	ω_N	−1.37(6)	−2.3(11)	8.2(12)	−3.9(12)
	ω_R	2.06(3)	1.6(8)	−2.6(10)	−4.1(7)
Rb	ω_N	4.45(6)	6.6(10)	−1.9(12)	1.8(11)
	ω_R	−1.27(4)	−1.0(10)	−1.6(11)	1.7(10)
Cs	ω_N	−6.93(7)	2.6(13)	1.0(14)	−1.0(14)
	ω_R	−2.90(6)	3.8(10)	8.5(13)	5.0(10)

The numerical values of the susceptibilities $\chi^{(n)}(-n\omega)$ for specific atoms are governed by matrix elements of complicated form [see Eqs. (4) and (5)] and depend strongly on the frequency ω as well as on the structure of the atomic spectrum. Tables I and II give the calculated values of $\chi^{(5)}(-5\omega)$ for a number of atoms at the frequencies of neodymium ($\omega_N = 9440$ cm^{-1}) and ruby ($\omega_R = 14\,400$ cm^{-1}) lasers. (The details of the calculations are given in the Appendix.) For comparison, these tables include also the susceptibilities $\chi^{(3)}(-3\omega)$ (Ref. 5). It is clear that at a fixed value of ω the tabulated susceptibilities $\chi^{(n)}$ rise rapidly on increase in n. This is demonstrated more clearly by the values of $\chi^{(n)}(-n\omega)$ for the hydrogen atom at a frequency ω_N (in atomic units): $\chi^{(1)} = 4.55$; $\chi^{(3)} = 2.79 \times 10^2$; $\chi^{(5)} = 9.14 \times$

TABLE II. Nonlinear susceptibilities $\chi^{(3)}(-3\omega)$ and $\chi^{(5)}(-5\omega)$ for generation of third and fifth harmonics in rare gases (values of χ are given in atomic units).

Atom	ω_N		ω_R		$2\omega_N$	
	$\chi^{(3)}(-3\omega)$	$\chi^{(5)}(-5\omega)$	$\chi^{(3)}(-3\omega)$	$\chi^{(5)}(-5\omega)$	$\chi^{(3)}(-3\omega)$	$\chi^{(5)}(-5\omega)$
Ne	45.0	2.1(3)	46.0	3.2(3)	54.8	6.3(3)
Ar	390	6.9(4)	420	2.1(5)	560	−6.7(6)
Kr	870	2.5(5)	1.2(3)	1.5(6)	1.3(3)	1.0(6)
Xe	2270	1.6(6)	2.5(3)	−9.5(6)	5.6(3)	−1.8(8)

10^4; $\chi^{(7)}=2.19\times10^8$. Such very rapid rise of the nonlinear coefficients of atoms on increase in the nonlinearity n has been observed earlier for other many-photon processes in atoms.[11]

Since $\chi^{(n)}\gg\chi^{(n-2)}$, we can have situations in which generation of higher harmonics in sufficiently strong fields is more effective than the generation at the frequency 3ω. The results of Tables I and II allow us to investigate this process quantitatively and to identify the range of fields F_{opt} characterized by $\sigma(5\omega)>\sigma(3\omega)$, where F_{opt} is still less than the critical field resulting in breakdown of the medium as a result of many-photon ionization. This situation is possible in rare gases characterized by high ionization potentials and it has been observed experimentally in the generation of harmonics of the frequency $\omega\approx37\,600$ cm^{-1} ($4\omega_N$) in helium atoms.[12] It is reported that for a peak intensity $I_\omega^{peak}\approx10^{15}$ W/cm^2 of the incident radiation the ratio of the intensities of the fifth and third harmonics reached $I_{5\omega}/I_{3\omega}\approx20$. Clearly, the space and time inhomogeneities of the incident radiation will reduce the effective value I_ω^{eff} to somewhat less than I_ω^{peak}. Using Eq. (7) and the numerical values of $\chi^{(n)}$ for He at the frequency $\omega=4\omega_N$ from Table I (including fifth-order corrections to $\chi^{(3)}$) we obtain $I_{5\omega}/I_{3\omega}=22$ for $I_\omega=3.5\times10^{14}$ W/cm^2, which is in agreement with the experimental results of Ref. 12. For other atoms the inequality $I_{5\omega}>I_{3\omega}$ is obeyed at much lower intensities I_ω. For example, in xenon at the frequency $\omega=2\omega_N$ this inequality is satisfied when $I_{2\omega_N}\approx1.5\times10^{12}$ W/cm^2. In the case of picosecond pulses the probability of six-photon ionization of Xe in such a field is negligible ($W_{ion}^{(6)}\leq10^9$ sec^{-1}, Ref. 13) so that efficient generation of higher harmonics in Xe is also fully attainable.

The situation is different in alkali metal vapors. Although the condition $I_{5\omega}>I_{3\omega}$ is satisfied at even lower pump intensities (for example, in the case of cesium at $I_{\omega_N}\approx10^{11}$ W/cm^2), but alkali atoms are rapidly ionized in fields of this kind (the probability of four-proton ionization of cesium at $\omega=\omega_N$ is $W_{ion}^{(4)}=0.7\times10^{12}$ sec^{-1}, Ref. 13). This is true also of other alkali atoms so that we can see that in the case of atoms with low ionization potentials the generation of higher harmonics cannot be more efficient than the generation of the third harmonic. This is supported also by the experimental results on the generation of harmonics in sodium atoms,[14] where in addition to generation of the frequency 3ω there have also been generation at frequencies 5ω and 7ω, but the efficiency of this process decreases rapidly on increase in n.

Earlier numerical estimates of $\chi^{(5)}(-5\omega)$ for alkali atoms have been carried out only for $\omega=\omega_N$ and are given in Ref. 7 (all alkali atoms), Ref. 15 (Rb atom), and Ref. 2 (Na atom). In all these cases the calculations are based on allowance for a finite number of terms in the sums over intermediate states (Green functions G_E) in the matrix elements of Eq. (5) and they differ only in the number of the terms which are included. In the case of rubidium we have $\chi^{(5)}(-5\omega_N)=1,82\cdot10^{11}$ atomic units (a.u.) according to Ref. 15 and -1.98×10^{10} a.u. according to Ref. 7. Our calculation ($\chi^{(5)}=5.65\times10^{10}$ a.u.) is in good agreement with the most accurate estimates.[15]

In the case of sodium, we have $\chi_{Na}^{(5)}(-5\omega_N)=9.7\cdot10^{10}$ a.u. according to Ref. 2 and -3.91×10^9 a.u. according to Ref. 7. Our result is $\chi^{(5)}=1.02\times10^{10}$ a.u. The experimental value $\approx(2.5\pm1.25)\times10^9$ a.u. is given in Ref. 14.

§3. INFLUENCE OF HIGHER-ORDER EFFECTS ON NONLINEAR SUSCEPTIBILITIES

The vector $\mathbf{P}^{(n)}(n\omega)$ in Eq. (6) is only the first term in the expansion of the vector $\mathbf{P}(n\omega)$ in Eq. (1) as a series in powers of F. Since high-intensity fields are used in experiments (particularly to generate higher harmonics) the corrections to $\mathbf{P}^{(n)}(n\omega)$ governing the deviations from the power law

$$I_{n\omega}\propto(I_\omega)^n \tag{8}$$

in the generation of the n-th harmonic can be very important. The practical aspect is whether these corrections can reach a considerable value and whether they are observable in the range of three breakdown fields when the ionization effects are still unimportant.

In the range of validity of perturbation theory the main interest lies in the first correction term of the expansion

$$\mathbf{P}(n\omega)=\mathbf{P}^{(n)}(n\omega)+\mathbf{P}^{(n+2)}(n\omega)+\ldots,$$

which is governed by the nonlinear susceptibility $\chi_{ij_1\ldots j_{n+2}}^{(n+2)}$:

$$P_i^{(n+2)}(n\omega)=F\left(\frac{F}{2}\right)^{n+1}\sum_{j_1\ldots j_{n+2}}\chi_{ij_1\ldots j_{n+2}}^{(n+2)}(-n\omega;\underbrace{-\omega,\omega,\ldots,\omega}_{n+1})e_{j_1}^*e_{j_2}\ldots e_{j_{n+2}} \tag{9}$$

Here, $\chi^{(n+2)}(-n\omega)$ gives rise to linear (in respect of the intensity) corrections to $\chi^{(n)}(-n\omega)$. It is important to note that in Eq. (9) we have not only e_j but also the projections e_j^*, so that

$$\mathbf{P}^{(n+2)}(n\omega)=(I_\omega)^{(n+2)/2}\{A\mathbf{e}+B\mathbf{e}^*\}, \tag{10}$$

(A and B are the combinations of the components $\chi^{(n+2)}$) and for a nonzero degree of circular polarization $\mathbf{F}(t)(\mathbf{e}\neq\mathbf{e}^*)$ the correction terms make the polarization of the harmonics different from the polarization of the pump field [see Eq. (6)].

For an atom in the S state the tensor $\chi_{ij_1\ldots j_{n+2}}^{(n+2)}$ has two linearly independent components

$$\chi_{xx\ldots x}^{(n+2)}(-n\omega)\equiv\chi_\parallel,\quad \chi_{yyx\ldots x}^{(n+2)}(-n\omega)\equiv\chi_\perp,$$

which (in the static limit) are related by the general expression applicable to all nonlinear processes:

$$\chi_\parallel=(n+2)\chi_\perp|_{\omega=0}.$$

In contrast to Eq. (4), the quantum-mechanical expression for $\chi^{(n+2)}$ is much more cumbersome and contains terms of three types. There are $(n+3)(n+2)$ matrix elements of the $(n+2)$-th order given by Eq. (5), corresponding to the graphs in Fig. 1 with all possible transpositions of the photon lines. Apart from these terms, $\chi_\parallel$ contains the "normalization" and "circular" terms,[16] which do not appear in the first nonvanishing

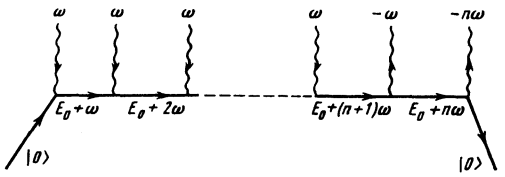

FIG. 1.

order of perturbation theory. In view of the cumbersome nature of the final formulas for $\chi_{\perp,\parallel}$ they will not be given here but they can be obtained in the same way as the higher-order corrections to the magnetoelectric susceptibilities.[16]

We shall now consider in greater detail the correction to the susceptibility $\chi^{(3)}(-3\omega)$, corresponding to the generation of a third harmonic.[1)] Nonzero components of the tensor $\chi^{(5)}(-3\omega)$ can be expressed in terms of $\chi_\parallel$ and $\chi_\perp$:

$$\chi^{(5)}_{yyyzzz}(-3\omega)=\chi^{(5)}_{zyyyzz}(-3\omega)=\frac{1}{4}(\chi_\parallel-\chi_\perp),$$

$$\chi^{(5)}_{zzyyzz}(-3\omega)=\frac{1}{6}(\chi_\parallel+\chi_\perp).$$

Transposition of the last four indices or relabeling of the indices does not alter the value of the component. Therefore, all the nonzero components can be obtained from the above relationships and Eq. (10) for $n=3$ can be rewritten in the form

$$\mathbf{P}^{(5)}(3\omega)=\frac{lF^5}{16}[\mathbf{e}(\chi_\parallel-\chi_\perp)+\mathbf{e}^*l\chi_\perp]. \tag{11}$$

Hence, the intensity-dependent degree of ellipticity of the polarization of the harmonic $\xi(F)=iP_y(3\omega)/P_x(3\omega)$ given by

$$\xi(F)=\xi\left\{\chi^{(3)}(-3\omega)+\frac{F^2}{4}(\chi_\parallel-\chi_\perp-l\chi_\perp)\right\}\Big/\left\{\chi^{(3)}(-3\omega)+\frac{F^2}{4}(\chi_\parallel-\chi_\perp+l\chi_\perp)\right\} \tag{12}$$

or, assuming that $|\mathbf{P}^{(5)}(3\omega)|\ll|\mathbf{P}^{(3)}(3\omega)|$,

$$\xi(F)\approx\xi\left(1-\frac{1}{2}lF^2\chi_\perp/\chi^{(3)}(-3\omega)\right).$$

Numerical values of the components $\chi_{\parallel,\perp}$ for alkali atoms, hydrogen, and helium calculated by the same method as $\chi^{(5)}(5\omega)$ (see Appendix) are given in Table I. The values of $\chi_\parallel$ and $\chi_\perp$ have a much more complicated frequency dependence than $\chi^{(4)}(-3\omega)$. In particular, $\chi_{\parallel,\perp}$ have resonance singularities at the frequency 4ω for the intermediate S and D states, as well as ω and 3ω resonances for the F states, which are not observed in the frequency dependence of $\chi^{(3)}(-3\omega)$. Then, in the vicinity of four-proton resonances of the S states we have $\chi_\parallel\approx\chi_\perp$, whereas for the D states and for the 3ω resonances of the F states we obtain $\chi_\parallel\approx-2\chi_\perp$, and in the vicinity of the ω resonances of the F states we have $\chi_\parallel\approx\frac{3}{2}\chi_\perp$. Moreover, in the case of $\chi_{\parallel,\perp}$ there is the possibility of the ω and 2ω resonances of the second order of the P and D states, respectively, and of the ω resonances of the third order of the P states [$\chi^{(3)}(-3\omega)$ and $\chi^{(5)}(-5\omega)$ have only first-order resonances].

In the vicinity of multiple resonances the relationship between $\chi_\parallel$ and $\chi_\perp$ depends on the structure of the spectrum of a specific atom. We can expect simultaneous appearance of several types of resonance, which increases considerably $P(3\omega)$ and enhances or weakens stimulated emission, depending on the relative signs of the quantities $\chi^{(3)}(-3\omega)$ and $\chi_{\parallel,\perp}$. It should be pointed out that an estimate of the component $\chi_\parallel$ for Rb at the frequency $\omega_N(\chi_\parallel=4.54\times10^{12}$ a.u.) obtained by Puell *et al.*[15] is close to our result (Table I) in respect of the absolute magnitude but opposite in sign.

The results of Table I allow us to analyze some features of the dependence of the efficiency of third harmonic generation on the pump intensity I_ω, and to find the limits of validity of the calculations of $\chi(-3\omega)$ by perturbation theory in the case of strong fields. The presence of second- and third-order resonance singularities in the frequency dependence of $\chi_{\parallel,\perp}$ has the effect that in the optical range the susceptibilities of alkali atoms obey $|\chi_{\parallel,\perp}|\gg|\chi^{(5)}(-5\omega)|$. This means that the change in $\chi(-3\omega)$ due to the higher-order effects appears in fields of lower intensity than those necessary to satisfy the condition $I_{5\omega}>I_{3\omega}$. A comparison of $\chi_{\parallel,\perp}$ with $\chi^{(3)}(-3\omega)$ shows that the nonlinear corrections become important in fields $F\gtrsim10^6$ V/cm. In these fields the ionization (at least in the case of picosecond pulses) does not yet result in breakdown and the nonlinear corrections to $\chi^{(3)}(-3\omega)$ may be manifested experimentally.

An experimental investigation of the generation of the third harmonic of high-power neodymium laser radiation in Rb and Na vapors was reported in Refs. 15 and 18. Deviation of the intensity $I_{3\omega_N}$ from the law (8) was observed for Rb in fields $I_\omega\gtrsim10^{10}$ W/cm^2 (Ref. 15) and for Na in fields $I_\omega\gtrsim3\times10^{11}$ W/cm^2 (Ref. 18). In both cases the rise of $I_{3\omega_N}$ slowed down when I_{ω_N} was increased and this was attributed to the influence of nonlinear effects (a change in the refractive index of Rb and many-photon ionization of Na) on the phase matching condition. Allowance for the fifth-order corrections to $\chi^{(3)}(-3\omega)$ also could reduce the third harmonic yield because the signs of $\chi^{(3)}$ and $\chi_\parallel$ (the experiments were carried out in linearly polarized fields) were opposite (Table I). However, in the case under consideration the effect should be slight because at the indicated intensities the change in $\chi^{(3)}$ would amount to just a few percent and become important only at intensities I_ω an order of magnitude higher. It should be noted that the change in $\chi^{(3)}$ for other atoms may be considerable even in weaker fields. For example, in the case of cesium atoms a change in $\chi^{(3)}$ amounts to 50% in a ruby laser field with $I_\omega\approx5\times10^9$ W/cm^2.

§4. CONCLUSIONS

We have considered two questions. 1) We have discussed the relationship between the nonlinear susceptibilities governing nonresonant generation of the third and fifth harmonics of optical radiation in atomic gases. We have assumed that the field is sufficiently weak so that only the first nonvanishing order of perturbation theory need be considered in calculations of the relevant atomic characteristics. 2) We have also analyzed the role of the nonlinear corrections to the susceptibility which are important in strong fields and which result

in deviations of the yield of a given harmonic from the power law (8).

The role of the nonlinear corrections is very important in the experimental generation of harmonics in gases employing strong fields. The change in the functional dependence (8) with an increase in I_ω may be due to various factors: a) a change in the conditions of propagation of the generated radiation (for example, failure of phase matching because of a nonlinear change in the refractive index or depletion of the nonlinear medium because of the ionization of the atoms); b) reaction of the generated radiation on the nonlinear polarization of the medium, which may occur when the intensities are high so that $I_{3\omega} \lesssim I_\omega$; c) a change in the nonlinear susceptibility of atoms in a strong pump field. In contrast to the first two, the last factor is not an effect of the medium but it is due to the action of the field on an isolated atom.

Effects of the b) and c) types have been analyzed earlier for specially selected conditions in intermediate resonances (see, for example, the recent work of Melikyan and Saakyan[19]). In this case the analysis simplifies greatly because all the effects are due to the field-induced perturbation of a local group of resonance levels, which can be allowed for quite satisfactorily by the zeroth approximation of the resonance perturbation theory.[17] The actual structure of the atom is unimportant and it is manifested only in the parametric dependence of the polarization vector on the dipole moments of resonance transitions.

We have considered nonlinear corrections to $\chi^{(n)}(-n\omega)$ in a typical off-resonance situation where the change in $\chi^n(-n\omega)$ is due to the field-induced change in the properties of atoms as a whole and this change cannot be attributed to a perturbation of any particular level (or a group of levels). It follows from the results of Table I that the corrections are compared with the main term in fields $F_{cr} \gtrsim (2-5)\times 10^6$ V/cm and, consequently, at $F \sim F_{cr}$ we need to consider specially the problem of the possibility of expanding $\mathbf{P}(n\omega)$ as a series in F. It is interesting to note that off-resonance corrections do not necessarily result in saturation-slowing down of the rise of $I_{n\omega}$ on increase in I_ω-but they may also enhance the effect, depending on the relative signs of $\chi^{(3)}$ and $\chi_{\parallel,\perp}$ (Table I). Promising results can be expected for experiments carried out in a strong field of elliptically polarized radiation, because it follows from Eq. (12) that in this case the change in the ellipticity of the polarization of the harmonic will make it possible to separate reliably the contribution of the nonlinear corrections to $\chi^{(n)}$ (and at the same time determine the relative signs of χ^n and $\chi_1^{(n+2)}$).

Clearly, under off-resonance conditions the value of F_{cr} is considerably higher than at resonances (for example, in the case of alkali atoms, we have $F_{cr}^{res} \lesssim 10^5$ V/cm). However, in the case of ultrashort pulses the possibilities of experimental observation of off-resonance generation of harmonics in fields $F \sim F_{cr}$ are not limited to the effects of dissociation of atoms as a result of ionization. Therefore, such experiments can give information on nonlinear characteristics of atoms in the range of fields, where the traditional (for nonlinear optics) scheme of describing off-resonance effects in terms of nonlinear susceptibilities independent of F is no longer valid.

APPENDIX. Details of Numerical Calculations of $\chi^{(m)}(-k\omega)$ Tensors

A numerical calculation of $\chi^{(m)}(-k\omega)$ reduces to a calculation of the composite matrix elements $T(\omega_1, \ldots, \omega_n)$ of the type given by Eq. (5). In the calculation of T an allowance is made for the interaction of the field with just the outer (optical) electron which moves in the effective potential of the nucleus and of the core atoms. Integration with respect to angular variables is carried out using the algebra of angular momenta. In the case of rare gases the separation of the angular variables is made in the jl-coupling scheme allowing for two branches of the excitation of the valence electrons corresponding to different momenta j_0 of the core ($j_0 = 1/2, 3/2$). (Some details of such calculations can be found in Ref. 13.) The resultant expression for T contains a combination of the radial integrals of the type

$$\rho_{l_1,\ldots,l_n}(\omega_1,\ldots,\omega_n) = \langle 0|r_1 g_{l_1}(E_0+\omega_1; r_1, r_2) r_2 \ldots r_n g_{l_n}(E_0+\omega_n; r_n, r_{n+1}) r_{n+1}|0\rangle,$$

where $g_l(E; r, r')$ is the radial Green function described for $E<0$ by an expansion as a series in terms of a complete set of the Sturm functions using the approximation of the model potential method.[5] This expansion does not include integration over the continuous spectrum and it makes it possible to use a simple algorithm for numerical calculation of ρ corresponding to all the values of the degree of nonlinearity n of practical interest.

A detailed account of the procedure used in numerical calculations for arbitrary values of n can be found in Ref. 20. If $E>0$, the Sturm expansion of gl is unsuitable and gl is described by integral representations or by a spectral expansion. Calculations carried out for the hydrogen atom (see Ref. 17) indicated that the contribution of the integral over the continuous spectrum in the spectral expansion of gl for $E>0$ is usually slight. Therefore, when $\rho(\omega_1, \ldots, \omega_n)$ contains the Green functions $gl(E; r, r')$ with $E>0$, we allowed only for the contributions of states in the discrete spectrum, which required a slight modification of the calculation procedure compared with that described earlier.[20]

[1)] It should be noted that $\chi_{ijkl}^{(3)}(-\omega)$ gives a F^2-dependent correction to the refactive index at a frequency ω and it also determines the dynamic hyperpolarizability of an atom in a field with elliptic polarization, and this aspect has been studied in detail earlier.[11, 17]

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Birefringence and gyrotropy due to nearly Bragglike processes in the x-ray region

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Propagation of electromagnetic waves is considered in scalar spatially periodic media at angles and frequencies that almost satisfy the Bragg conditions. The process of virtual rescattering into other waves and back leads to corrections to the phase velocity of the initial wave. The dependence of the amplitude of the scalar scattering on the polarization causes these corrections to produce birefringence in the region of the two-wave Bragg resonance. Near three-wave and multiwave resonances, subject to definite conditions on the symmetry of the medium, these corrections can lead also to gyrotropy, i.e., to rotation of the plane of polarization of the wave. The possibility of observing the effects in crystal at x-ray frequencies is considered.

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1. INTRODUCTION

It is known that the dielectric constant of a condensed medium differs from unity in the x-ray band ($\lambda \sim 1$ Å) only in the fourth or even fifth decimal point, and is furthermore a pure scalar:

$$\varepsilon_{ik}(\mathbf{r}) = \delta_{ik}\left[1 - \frac{4\pi N(\mathbf{r})e^2}{m\omega^2}\right],$$

where m and e are the mass and charge of the electron, $N(\mathbf{r})$ is their density, and $\omega = 2\pi c/\lambda$ is the radiation frequency. The propagation of x-ray photons through a medium is therefore not accompanied as a rule by a change in their polarization state.

Exceptions are cases when the conditions of Bragg rescattering from a given wave into another at the corresponding Fourier component $e_{\mathbf{q}}\exp(i\mathbf{q}\cdot\mathbf{r})$ of the dielectric constant are satisfied in the crystal. Since the amplitude of the scattering from a wave A into a wave B on scalar perturbations is proportional to $f_{BA} \propto e_{\mathbf{q}}(\mathbf{e}_B^*\cdot\mathbf{e}_A)$, where $\mathbf{e}_A$ and $\mathbf{e}_B$ are the unit vectors of the polarization, it follows, as is well known, that the Bragg interaction is different for the s- and p-polarizations in both the kinematic and the dynamic theory (see, e.g., Refs. 1–3). More complicated are the polarization effects in the case of multiwave refraction (see Refs. 1–4). In all these cases, however, apart from the change in the polarization state of the incident wave itself, diffracted waves are excited in fact.

We wish to discuss in this paper the possibility of observing birefringence and gyrotropy in pure form, i.e., without real excitation of other waves. As the mechanism for producing these effect we propose the process of virtual rescattering into other waves and back. The smallness of the amplitude of the elementary rescattering act can be offset to a considerable degree by the proximity to the Bragg resonances.

2. BIREFRINGENCE IN PROPAGATION NEAR A SOLITARY BRAGG RESONANCE

We consider wave propagation in a direction close to the satisfaction of the Bragg condition for the Fourier component of the dielectric constant

$$\delta\varepsilon(\mathbf{r}) = 2|\delta\varepsilon_{\mathbf{q}}|\cos(\mathbf{qr}+\varphi) = \delta\varepsilon_{\mathbf{q}}e^{i\mathbf{qr}} + \delta\varepsilon_{-\mathbf{q}}e^{-i\mathbf{qr}}. \quad (1)$$

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