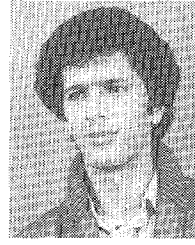




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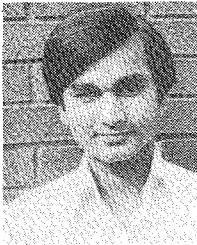
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Large dc-Electric-Field-Induced Nonlinear Optical Susceptibility of Hydrogen Atomic Vapors

ROBERT W. BOYD AND LIAN-QIN XIANG

Abstract—A unique set of circumstances makes the Rydberg states of atoms ideal for mediating tunable, resonantly enhanced, nonlinear optical interactions. The application of a dc electric field to a vapor of such atoms can induce a second-order or $\chi^{(2)}$ nonlinear susceptibility, which in the absence of the applied field would be forbidden by reasons of symmetry. This paper presents an exact expression, valid within the hydrogenic approximation for field strengths that produce a linear Stark effect, for the resonant contribution to the dc-electric-field-induced $\chi^{(2)}$ susceptibility. Numerical evaluation of this expression for field strengths of the order of 10^3 V/cm predict $\chi^{(2)}$ susceptibilities hundreds of times larger than those of conventional mixing crystals.

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I. INTRODUCTION

THE Rydberg levels of atoms are characterized by several features, such as large electric polarizabilities and large electric dipole transition moments, that make them desirable systems in which to study nonlinear optical interactions. It is well known, however, that the lowest order, or $\chi^{(2)}$, nonlinearity of a free atomic vapor must vanish by reasons of symmetry. The application of a dc electric field can break the inversion symmetry, thus allowing the $\chi^{(2)}$ nonlinearity to exist [1]–[4]. This paper derives within the hydrogenic approximation a closed-form expression for the magnitude of the resulting optical nonlinearity. It is found that fields of only modest strength are required to produce large values of this induced $\chi^{(2)}$ susceptibility.

It is intended that the calculation presented here, which is exact over a wide range of field strengths for the hydrogen

atom, should approximately predict the magnitude of the $\chi^{(2)}$ nonlinearity for the highly excited states of any atomic system. These highly excited levels are known as Rydberg levels because their energies obey (usually with the inclusion of a quantum defect) the Rydberg energy formula. These levels are connected by large dipole moments, possess long radiative lifetimes, and display a large Stark effect. These characteristics make the Rydberg levels suitable for mediating tunable, resonantly enhanced nonlinear optical processes. Laser action between Rydberg levels has been studied by Grischkowsky *et al.* [5], four-wave mixing has been studied by Sorokin *et al.* [6], and Stark tunable detection of infrared radiation has been demonstrated by Gelbwachs *et al.* [7]. By the application of a dc electric field to an atomic vapor, it should be possible to study Stark-tunable resonantly enhanced mixing processes involving the atomic Rydberg levels.

II. THEORETICAL FORMULATION

Fig. 1 illustrates the process considered in this paper. Optical fields at frequencies $\omega_1 \simeq \omega_{ji}$, $\omega_2 \simeq \omega_{kj}$, and $\omega_3 = \omega_1 + \omega_2$ interact via the nonlinear response of an atomic vapor. This process can be described by the nonlinear susceptibility [8]

$$\chi^{(2)} = \frac{N\mu_{ik}\mu_{kj}\mu_{ji}}{2\hbar^2(\omega_{ji} - \omega_1)(\omega_{ki} - \omega_2 - \omega_1)} \quad (1)$$

where N denotes the number density of atoms and μ_{lm} denotes the electric dipole matrix element connecting level l with level m . Equation (1) is valid as long as the frequency detunings appearing in the denominator are sufficiently small that the contributions from other atomic levels can be neglected, yet are much larger than the linewidths of the atomic transitions and of the optical fields.

If each of the levels i , j , and k have definite parity, at least one of the μ_{lm} 's will be zero, causing $\chi^{(2)}$ to vanish. The application of certain perturbations, such as a dc electric field, can mix levels having opposite parity, thus allowing each of the μ 's to be nonzero. In principle, it is possible to calculate the dc-field-induced μ_{lm} for any atomic system by first calculating the eigenstates of the atom in the presence of the perturbation, perhaps by using perturbation theory or by diagonalizing the matrix of the perturbing Hamiltonian. We have chosen instead to work in the hydrogenic approximation, for which case the use of parabolic coordinates leads to closed-form expressions for the matrix elements. This closed-form solution is valuable both because it provides a basis for understanding the underlying physical processes and because it should roughly predict the size of the matrix elements for the Rydberg states of atoms other than hydrogen.

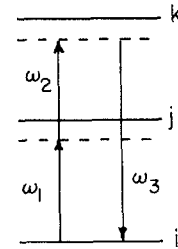


Fig. 1. Optical waves with frequencies ω_1 , ω_2 , and $\omega_3 = \omega_1 + \omega_2$ interact via the nonlinear response of an atom with energy eigenstates labeled i , j , and k .

To lowest order in the applied electric field strength E , and neglecting fine-structure corrections, the energy eigenvalues of hydrogen are given by [9]

$$W = \frac{-1.097 \times 10^5}{n^2} + \frac{E}{15620} n(n_1 - n_2) \quad (2)$$

where E is measured in V/cm, W is measured in cm^{-1} , and where the principal quantum number n is related to the magnetic quantum number m and to the electric quantum numbers n_1 and n_2 , obtained by solving the hydrogenic Schrodinger equation in parabolic coordinates by

$$n = n_1 + n_2 + |m| + 1. \quad (3)$$

Fine structure splittings are negligible compared to Stark splittings whenever

$$\frac{\alpha^2}{6n^3} \ll \frac{E}{5.142 \times 10^9} \quad (4a)$$

where $\alpha = e^2/\hbar c$ is the fine-structure constant, while the quadratic Stark splittings are negligible compared to linear Stark splittings whenever [9]

$$\frac{E}{5.142 \times 10^9} \ll \frac{24}{n^3(17n^3 + 19)}. \quad (4b)$$

These inequalities are satisfied for field strengths roughly in the range 10^3 – 10^4 V/cm for principal quantum numbers in the range 1–10.

Closed-form expressions for the electric-dipole matrix elements connecting the energy eigenstates $(n_1 n_2 m)$ and $(n'_1 n'_2 m')$ can be obtained when the inequalities (4) are satisfied. The only selection rule that has to be satisfied in order for the matrix element to be nonzero is that $\Delta m = 0$ or ± 1 . For radiation polarized parallel to the direction of the dc electric field, the rule becomes $\Delta m = 0$ and the transition moment is given [10] for $m \geq 0$ by

$$\begin{aligned} \mu_{n_1 n_2 m}^{n'_1 n'_2 m} &= (-)^{n'_1 + n'_2} \frac{ea_0}{4(m!)^2} \sqrt{\frac{(n_1 + m)!(n_2 + m)!(n'_1 + m)!(n'_2 + m)!}{n_1!n_2!n'_1!n'_2!}} \left(\frac{4nn'}{(n - n')^2} \right)^{m+2} \left(\frac{n - n'}{n + n'} \right)^{n+n'} \\ &\times \left\{ \left[2(n'_1 - n'_2) \frac{n^2 + n'^2}{(n + n')^2} - (n_1 - n_2) \frac{4nn'}{(n + n')^2} \right] \Psi_m(n_1 n'_1) \Psi_m(n_2 n'_2) \right. \\ &\left. - 2[n'_1 \Psi_m(n_1, n'_1 - 1) \Psi_m(n_2, n'_2) - n'_2 \Psi_m(n_1, n'_1) \Psi_m(n_2, n'_2 - 1)] \right\} \quad (5) \end{aligned}$$

and for $m < 0$ is given by this expression with m replaced by $-m$. For radiation polarized perpendicular to the direction of the dc electric field, the selection rule becomes $\Delta m = \pm 1$, and the transition moment is given for $m \geq 1$ by

$$\begin{aligned} \mu_{n_1 n_2 m}^{n'_1 n'_2 m-1} &= \mu_{n'_1 n'_2 m-1}^{n_1 n_2 m} = (-)^{n'_1+n'_2} \frac{ea_0}{4(m-1)!^2} \\ &\cdot \sqrt{\frac{(n_1+m)!(n_2+m)!(n'_1+m-1)!(n'_2+m-1)!}{n_1!n_2!n'_1!n'_2!}} \\ &\cdot \left(\frac{4nm'}{(n-n')^2}\right)^{m+1} \left(\frac{n-n'}{n+n'}\right)^{n+n'} \\ &\times \left\{ \Psi_{m-1}(n_1 n'_1) \Psi_{m-1}(n_2 n'_2) - \left(\frac{n-n'}{n+n'}\right)^2 \right. \\ &\cdot \left. \Psi_{m-1}(n_1+1, n'_1) \Psi_{m-1}(n_2+1, n'_2) \right\} \quad (6) \end{aligned}$$

and for $m \leq -1$ is given by this expression with m replaced by $-m$ everywhere. In these formulas, e denotes the charge of the electron, $a_0 \equiv \hbar^2/me^2$ denotes the radius of the first Bohr orbit, and ψ denotes Gauss' hypergeometric function defined by

$$\psi_m(n_i, n'_i) = F\left(-n_i, -n'_i, m+1, \frac{-4nn'}{(n-n')^2}\right) \quad (7)$$

where

$F(\alpha, \beta, \gamma, x)$

$$= \sum_{\nu=0}^{\infty} \frac{\alpha(\alpha+1)\cdots(\alpha+\nu-1)\beta(\beta+1)\cdots(\beta+\nu-1)}{\gamma(\gamma+1)\cdots(\gamma+\nu-1)\nu!} x^\nu. \quad (8)$$

Since the allowed values of n_i and n'_i ($i = 1, 2$) are integers, the series given in (8) terminates. As a result of relations (5)–(8), the matrix elements must obey the following symmetry relations:

$$\mu_{n_1 n_2 m}^{n'_1 n'_2 m} = -\mu_{n_2 n_1 m}^{n'_2 n'_1 m} \quad (9)$$

and

$$\mu_{n_1 n_2 m}^{n'_1 n'_2 m-1} = \mu_{n_2 n_1 m}^{n'_2 n'_1 m-1}. \quad (10)$$

III. NUMERICAL EXAMPLES

The $\chi^{(2)}$ susceptibility for any set of energy levels can be obtained through use of (1) and (4)–(8). For simplicity, let us consider first the situation in which only states belonging to $n = 1, 2$, and 3 contribute to the nonlinear susceptibility. The energy-level structure in the presence of an applied field of magnitude E is shown in Fig. 2. In Table I we have listed all of the matrix elements that can contribute to the $\chi^{(2)}$ susceptibility. It can be noted that these values obey the relations (9) and (10).

From (1), the nonlinear susceptibility depends on the product of three matrix elements. This product has the form

$$\mu^{(2)} = \mu_{n'_1 n'_2 m}^{n_1 n_2 m} \mu_{n'_1 n'_2 m'}^{n_1 n'_1 n'_2 m''} \mu_{n_1 n_2 m}^{n'_1 n'_2 m'}. \quad (11)$$

The fifteen possible values of $\mu^{(2)}$ are tabulated in Table II in units of $e^3 a_0^3$. For an appropriate choice of intermediate levels,

m =	-2	-1	0	1	2
n = 3	$\frac{00-2}{01-1}$	$\frac{10-1}{01-1}$	$\frac{200}{020}$	$\frac{101}{011}$	$\frac{002}{011}$
n = 2	$\frac{00-1}{010}$	$\frac{100}{010}$	$\frac{001}{010}$		
n = 1			$\frac{000}{000}$		

Fig. 2. Energy level structure of hydrogen in the presence of a dc electric field. Energy eigenstates are labeled by the quantum numbers (n_1, n_2, m) obtained by solving Schrodinger's equation in parabolic coordinates.

$\mu^{(2)}$ is of order unity, suggesting the possibility of a large optical nonlinearity. The entries in Table II can be divided into four classes which depend upon the polarizations of the interacting fields, as illustrated schematically in the inset to the table. If the lasers are detuned from resonance by an amount much greater than the Stark splitting of the various levels, the $\chi^{(2)}$ susceptibility becomes proportional to the sum of all of the $\mu^{(2)}$'s for the appropriate polarizations. It can be noted that the terms within each class in Table II sum to zero. Thus, if the lasers cannot resolve the Stark splitting of the atom, the $\chi^{(2)}$ susceptibility becomes zero, as in the case of an atom in an isotropic environment.

Our first example (involving the levels $n = 1, 2$, and 3) was chosen for its simplicity, and we were thus able to tabulate all of the possible values of $\mu^{(2)}$. However, the $n = 1$ to $n = 2$ transition of hydrogen occurs at a wavelength of 1215 Å, where no laser is presently available. We can consider as a second example an interaction involving levels $n = 3, 8$, and 11. For this example, ω_1 corresponds to a frequency of $\sim 10\,000$ cm^{-1} , ω_2 to a frequency of ~ 900 cm^{-1} , and ω_3 to a frequency of $\sim 11\,000$ cm^{-1} . This mixing process could thus be used to detect an infrared signal at ω_2 by the process of upconversion or, assuming that ω_1 and ω_3 are the applied frequencies, it could be used to generate tunable infrared radiation by the process of difference-frequency generation. In either case, tunability can be obtained by varying the strength of the applied electric field. For our example, the largest tuning rates occur for the $(12, 0, 0)$ and $(0, 12, 0)$ states of the $n = 13$ manifold, and are equal to 0.01 $\text{cm}^{-1}/(\text{V}/\text{cm})$. It is assumed that population is maintained in the $n = 3$ state (if it is not the ground state of the atom in question) by optical pumping.

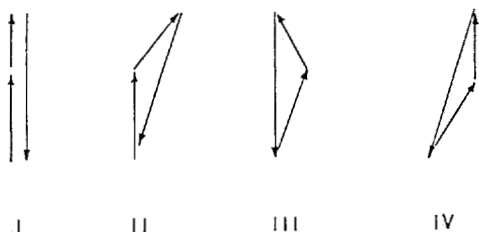
We have calculated all 890 of the matrix elements that can appear in the expression for $\chi^{(2)}$, and have determined numerically that the largest value of $\mu^{(2)}$ occurs for the interaction involving the levels $(2, 0, 0)$, $(7, 0, 0)$, and $(12, 0, 0)$ and is equal to $-0.1788e^3 a_0^3$. The $\chi^{(2)}$ nonlinearity can be estimated by assuming an initial lower state population of 1×10^{14} atoms/ cm^3 and equal detunings of 5 GHz. The value of $\chi^{(2)}$

TABLE I
 DIPOLE MATRIX ELEMENTS CONNECTING LEVELS $n_1 n_2 m$ AND $n_1' n_2' m'$

$\Delta m = 0$						$\Delta m = \pm 1$					
n	n_1	n_2	m	n'	μ	n	n_1	n_2	m	n'	μ
1	0	0	0	2	-0.5267	1	0	0	0	2	0.5267
	0	0	0	0	0.5267						
1	0	0	0	3	-0.2109	1	0	0	0	3	0.1492
	0	0	0	1	0						
	0	0	0	0	0.2109						
2	1	0	0	3	-1.8137	2	1	0	0	3	1.376
	1	0	0	1	1.194						
	1	0	0	0	-0.0442						
	0	0	1	1	-1.5014						
	0	0	1	0	1.5014						
	0	1	0	2	0.04424						
	0	1	0	1	-1.194						
	0	1	0	0	1.8137						

 TABLE II
 VALUES OF $\mu^{(2)} = \mu_{n_1 n_2 m, n_1' n_2' m'}^{(2)}$ FOR $n_1 = n_2 = m = 0$

$n_1' n_2' m'$	2 0 0	1 1 0	0 2 0	1 0 1	0 1 1
1 0 0	-0.2015	0.0	0.0049	-0.1081	0.0098
0 1 0	-0.0049	0.0	0.2015	-0.0098	0.1081
0 0 1	0.0147	0.0	-0.0147	-0.1180	0.1180



is then 1.33×10^{-7} , which is several hundred times greater than the nonlinear susceptibility of KDP.

IV. DISCUSSION

The large value of the $\chi^{(2)}$ susceptibility calculated in the last section should allow generation or detection of infrared radiation by nonlinear mixing to be a useful process. Since large $\chi^{(2)}$ susceptibilities are also predicted for other choices of principal quantum number, nonlinear optical mixing using Rydberg levels should also be efficient at other frequencies, including those in the far infrared where standard nonlinear mixing crystals are not transparent.

The resonant dc-field-induced $\chi^{(2)}$ nonlinearity discussed here is comparable in magnitude to that predicted and studied by Abrams *et al.* [4] for resonantly enhanced mixing in NH_2D in the presence of a dc electric field. Much smaller dc field-induced nonlinearities are achieved in nonresonant media. For the case of gaseous helium, Finn and Ward [3] have deter-

mined that the susceptibility for second-harmonic generation has the form

$$\chi^{(2)} = \chi^{(3)} E_0 \quad (12)$$

where E_0 is the magnitude of the dc electric field and where $\chi^{(3)}$ was measured to have the value 3.8×10^{-39} ESU/atom. For a dc field of 3000 V/cm, the resulting $\chi^{(2)}$ susceptibility is 10^{14} times smaller than the resonant value calculated in the last section. Bethea [4] has studied dc field-induced second-harmonic generation in glass and in liquid CH_2I_2 , and has measured values of $\chi^{(3)}$ of 48×10^{-14} ESU and 2.3×10^{-14} ESU, respectively. For a dc field of 3000 V/cm, these imply effective $\chi^{(2)}$ susceptibilities smaller than those predicted for atomic hydrogen by factors of $\sim 10^6$.

For atoms other than hydrogen, the theory developed here cannot provide quantitative predictions of the magnitude of the dc-field-induced $\chi^{(2)}$ susceptibility. Large $\chi^{(2)}$ susceptibilities would be expected, however, whenever the applied dc field is sufficiently large to produce significant mixing between states of opposite parity, thus inducing a linear Stark effect. Measurements involving the Rydberg states of atomic sodium have shown that field strengths of only ~ 100 V/cm are sufficient to produce such mixing [11]. It is necessary that the applied field strength not be so large as to lead to field ionization of the excited Rydberg state of principal quantum number n . It has been shown [12] that this breakdown field strength is of the order of the value $5.142 \times 10^9/n^4$ V/cm predicted by classical theory.

The theory developed in this paper has assumed that the three-wave mixing process can be described as a $\chi^{(2)}$ nonlinearity. For large optical field strengths, higher order effects can modify the predictions given here. However, the theory presented here will be valid as long as the ac Stark shifts [13] ($\sim |\mu E|$) of the atomic levels are much less than the laser detuning from resonance ($|\omega_{ij} - \omega|$) for each of the transitions in Fig. 1, and as long as population is not redistributed among the levels by the optical fields. The limitations imposed on the field strengths by the second condition are difficult to calculate exactly because of the complicated decay dynamics of Rydberg levels. It can be expected, however, by analogy to the exact results that are obtainable for a two-level atomic

system [14], that the second condition will be satisfied as long as

$$|\mu_{ij}E(\omega_1)| \ll \hbar |\omega_{ij} - \omega_1| (\delta\nu_{ij}\tau_j)^{1/2}$$

and

$$|\mu_{ik}E(\omega_3)| \ll \hbar |\omega_{ik} - \omega_3| (\delta\nu_{ik} - \tau_k)^{1/2}$$

where $\delta\nu_{ij}$ denotes the linewidth of the ij transition, τ_j denotes the lifetime of level j , and where the remaining symbols are defined in Fig. 1.

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Robert W. Boyd, for a photograph and biography, see this issue, p. 1207.



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Scalable Visible Nd:YAG Pumped Raman Laser Source

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Abstract—Experimental and analytical studies of a 60 Hz, 0.45 W, 630 nm Raman laser source with a 12 ns pulse duration have demonstrated an overall electrical to optical efficiency of 0.12 percent. This is the first demonstration of a short pulse, high repetition rate red laser at such a high average output power without the need for a visible pump laser. Additional significance arises from the fact that simple extensions of the present work will produce many wavelengths in the visible and near infrared (IR) spectral region. A 1064 nm Nd:YAG pump laser operating at 60 Hz was used to pump a methane gas Raman laser oper-

ating at 1544 nm. This wavelength was mixed with the remaining 1064 nm laser output in noncritically phase matched lithium niobate to produce 630 nm radiation. The optical energy conversion efficiencies for the three steps were 1.4, 30, and 20 percent, respectively, for output energies of 86 mJ at 1064 nm, 15 mJ at 1544 nm, and 7.5 mJ at 630 nm. rms pulse amplitude variation measured 6 percent or less. A 10.7 million pulse life test was conducted, and the average output energy did not vary more than ± 10 percent from its initial value.

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INTRODUCTION

HIGH pulse repetition frequencies (PRF's) at moderate average powers and efficiencies have been primary objectives of recent investigations of wavelength agile solid-state laser sources operating in the visible to near infrared spectral region. Nonlinear frequency conversion using stimulated Raman scattering [1]-[3] has been used with Nd:YAG lasers operat-