Quantum Electronics **30** (6) 520 – 522 (2000)

PACS numbers: 42.65.Ky; 78.20.Jq; 32.60.+i
DOI: 10.1070/QE2000v030n06ABEH001756

Optical Stark effect and Kerr nonlinearities of atomic media in nonlinear optical processes

I A Kulagin, T Usmanov

Abstract. It is shown that the resonant enhancements of the optical Kerr effect and the optical Stark effect of the same order are caused by the same change in the state of an atomic system. The analysis of the known experimental data demonstrates the identity of their influence on nonlinear optical processes. It is shown that a separate analysis of the influence of Stark shift and resonantly enhanced Kerr nonlinearities leads to incorrect results.

Among the main factors governing the interaction of optical radiation with atomic media are optical Kerr and Stark effects. These effects are commonly described by the perturbation theory, where the unperturbed atomic spectrum is taken as an initial basis and the action of the field is reduced to small corrections to the unperturbed energies of bound states [1 – 4]. In this case, one can expand the Stark shift of a level in a power series in the field, and the expansion coefficients represent susceptibilities of different orders [5]. In particular, the proportionality factor of the second-order shift of the ground level represents the linear polarisability of an atom, and the factor corresponding to the fourth-order shift represents a third-order Kerr nonlinearity. Note that the quadratic (electronic) Kerr effect is described by the third-order Kerr nonlinearities. Thus, these two effects are related to one another.

However, there exists a deeper interrelation, which is caused by a common nature of the Stark shift and the resonance parts of optical nonlinearities, in particular, Kerr nonlinearites. Nevertheless, in some papers, these two factors are treated to be independent, and the competition between these factors and cancelling out the effects caused by each of them is analysed (see, e.g., Ref. [6] and the references cited therein, and Refs [7, 8]). In this paper, we show that the optical Stark shift of levels of atomic states and their contribution to the Kerr nonlinearity of the same order are caused by the same change in the state of an atomic system, and we analyse the influence of these effects on the frequency conversion of laser radiation in gas media.

The response of an atomic system to the resonance interaction with the optical radiation $\mathscr{E}_1 = \{E_1 \exp[\mathrm{i}(\omega t - k_1 z)] + \mathrm{c.c.}\}/2$ can be described by the sum of resonance and nonresonance parts of polarisation [4 – 6]. The resonance part of polarisation is proportional to the element of the density matrix for the resonance transition σ_{n1}. One can describe the evolution of σ_{n1} using the generalised two-level system [4, 5]:

$$\frac{\partial \sigma_{n1}}{\partial t} + \mathrm{i}\left(\delta + \Omega_n - \frac{\mathrm{i}}{T_2}\right)\sigma_{n1} = \frac{\mathrm{i}}{\hbar}\eta V_n,$$
$$\frac{\partial \eta}{\partial t} + \frac{\eta - \eta_0}{T_1} = -\frac{4}{\hbar}\,\mathrm{Im}\left(\sigma_{n1} V_n^*\right), \tag{1}$$

where η is the difference of populations for the resonance transition; η_0 – is the equilibrium difference of populations; T_1 and T_2 are the longitudinal and the transverse relaxation times; $\delta = \omega_{n1} - n\omega$ is the frequency detuning from the resonance; the form of the perturbation V_n is determined by the resonance order n; and similarly, the parameter Ω_n of the Stark shift is determined by the order n of the approximation being used. In the third-order of the averaging method,

$$V_1 = \frac{1}{2} q_1 E_1 + \frac{1}{8} q_3 |E_1|^2 E_1, \quad V_2 = \frac{1}{4} q_2 E_1^2,$$

$$\Omega_2 = (4\hbar)^{-1}\left[\chi_1^{\prime(1)}(\omega;\omega) - \chi_n^{\prime(1)}(\omega;\omega)\right]|E_1|^2,$$

where $\chi_i^{(1)}(\omega;\omega)$ is the linear susceptibility of an atom in the state $|i\rangle$; q_n is the composite matrix element whose form is determined by the resonance order ($q_1 = \mu_{12}, q_2 = \hbar^{-1} \times \sum{}' \mu_{1i}\mu_{i3}(\omega_{i1}^{\mathrm{r}} - \omega)^{-1}$ etc. and $\omega_{ij}^{\mathrm{r}} = \omega_{ij} - \mathrm{i}/T_{ij}$; μ_{ij} and ω_{ij} are the dipole matrix element and the frequency of the transition between the states $|i\rangle$ and $|j\rangle$). The prime means that the resonance term is neglected. The solutions of (1) in the stationary case and in the adiabatic-following limit have the form (see, e.g., [6])

$$\sigma_{n1}^{\mathrm{s}} = \frac{[\mathrm{i} + (\delta + \Omega_n)T_2]T_2\eta_0 V_n}{1 + (\delta + \Omega_n)^2 T_2^2 + 4T_1T_2|V_n|^2}, \tag{2}$$

$$\sigma_{n1}^{\mathrm{a}} = \frac{[\mathrm{i} + (\delta + \Omega_n)T_2]\eta_0 V_n}{\left[(\delta + \Omega_n)^2 + 4|V_n|^2\right]^{1/2}(\delta + \Omega_n)T_2}. \tag{3}$$

The expressions for linear susceptibilities and the nonlinearities describing optical Kerr effect can be using methods of perturbation theory (see, e.g., [2 – 5, 9]):

$$\chi^{(1)}(\omega;\omega) = \hbar^{-1}\sum_i \mu_{1i}\mu_{i1}\left[(\omega_{i1}^{\mathrm{r}} - \omega)^{-1} + (\omega_{i1}^{\mathrm{r}} + \omega)^{-1}\right], \tag{4}$$

$$\chi^{(3)}(\omega;\omega,\omega,-\omega) = \hbar^{-3}\sum_{ijk}\mu_{1i}\mu_{ij}\mu_{jk}\mu_{k1}(C_{1i,1j,1k} - C_{i1,j1,k1}), \tag{5}$$

I A Kulagin, T Usmanov 'Akadempribor' Research and Production Association, Academy of Sciences of Uzbekistan, Akademgorodok, Tashkent, 700143 Uzbekistan; e-mail: kulagin@acpr.silk.org

Received 30 December 1999
Kvantovaya Elektronika **30** (6) 520 – 522 (2000)
Translated by A N Kirkin, edited by M N Sapozhnikov

where

$$C_{1i,1j,1m} = \left[(\omega^{\rm r}_{k1} - \omega)^{-1} + (\omega^{\rm r}_{k1} + \omega)^{-1}\right] \times (\omega^{\rm r}_{i1} - \omega)^{-1}(\omega^{\rm r}_{j1} - 2\omega)^{-1} \pm (1 - \delta_{1j}) \times \left[(\omega^{\rm r}_{k1} - \omega)^{-1} + (\omega^{\rm r}_{k1} + \omega)^{-1}\right](\omega^{\rm r}_{j1} \pm \omega)^{-1}/\omega^{\rm r}_{i1} + \delta_{1j}\left[(\omega^{\rm r}_{k1} - \omega)^{-1} + (\omega^{\rm r}_{k1} + \omega)^{-1}\right](\omega^{\rm r}_{i1} \pm \omega)^{-2}. \quad (6)$$

Because of the presence of secular terms in the derivation of (5), one should solve equations of form (1). This leads to the difference of (6) from the expressions obtained in the phenomenological way, this difference consisting in the appearance of the last term in the right-hand side of (6). From the analysis of (5), it follows that the Stark shift and the corresponding part of the Kerr nonlinearity are described by the same change in σ_{n1}. One can also see this by expanding (2) and (3) in a power series with accounting for (4)–(6) for a small shift $\Omega_n \ll \delta$ and a considerable detuning from resonance $\delta \gg 1/T_2$.

In the case of one-photon resonance, the first term of the expansion describes the resonance contribution to the linear susceptibility and the contribution made to the third-order Kerr nonlinearity, which is inversely proportional to the frequency detuning (higher-order terms in the averaging method describe the contribution to higher-order Kerr nonlinearities, which is also inversely proportional to frequency detuning). The second term of the expansion describes the contribution to the third- and fifth-order Kerr nonlinearities, which is inversely proportional to the square of frequency detuning. In this case, the shift of the excited and ground states corresponds to the contribution to the second and third terms of sum (6), respectively. One can similarly describe contributions to Kerr nonlinearities in the case of higher-order resonance.

Thus, in the case where it is required to take into account the Stark shift, one should exclude from the analysis the resonance contribution of the corresponding terms in all types of nonlinearities of different orders (both the nonlinearities describing the self-action of radiation and the nonlinearities determining the action of radiation fields on one another, for example, the nonlinearities of the form $\chi^{(3)}(\omega_i; \omega_i, \omega, -\omega)$, $\chi^{(5)}(3\omega; \omega, \omega, \omega, \omega, -\omega)$ etc.).

Ignoring this fact may lead to incorrect results. In particular, in Ref. [8], the authors experimentally and theoretically studied the generation of the third harmonic of a Nd : YLF laser in xenon near the three-photon resonance under conditions of strong focusing to the centre of the medium (the length of the medium L was much greater than the confocal parameter b). In this region, the medium has a positive dispersion and, as follows from the analysis, the harmonic should be absent because of the phase shift caused by focusing [6]. However, the third harmonic was experimentally observed, and its power dependence on the fundamental-radiation intensity was of an enormously high order. The analysis of the influence of optical Stark effect showed [8] that this effect, under the given experimental conditions, increased the frequency detuning from resonance, with the sign of dispersion being unchanged, and decreased the effective nonlinearity. From these facts, the conclusion was made that the optical Stark effect was not responsible for the frequency conversion and that the harmonic generation was caused by the fifth-order nonlinearity. Using expressions from Refs [10, 11] for the third-harmonic generation in focused beams on the basis of the fifth-order nonlinearity and the phenomenological description of the resonantly enhanced influence of the Kerr nonlinearity, an agreement was obtained between the experimental and the theoretical dependences of the harmonic signal on the xenon pressure. However, the calculation [10, 11] of the ratio between harmonic signals obtained at the optimum pressure under different focusing conditions gives the values that differ from the experimentally obtained ratio by a factor of more than 300. Thus, the analysis presented in Ref. [8] failed to describe the experiment, and the harmonic generation mechanisms remained unexplained.

Similarly to Ref. [12], we obtain the following expression for the amplitude of the third harmonic of the lowest mode of a Gaussian beam of fundamental radiation ($E_1(\xi, r) = E_{10}\exp\{-k_1 r^2/[b(1 + {\rm i}\xi)]\}/(1 + {\rm i}\xi)$, where $\xi = 2(z - f)/b$; E_{10} is the maximum beam amplitude at the waist; f is the beam waist position; z and r are the longitudinal and the transverse coordinates) under the conditions of three-photon resonance in the adiabatic-following limit:

$$E_3(\xi, r) = {\rm i}\frac{bu_1\gamma E_{10}^3}{2(1 + {\rm i}\xi)}\exp\left[-\frac{3k_1 r^2}{b(1 + \xi^2)} - \varepsilon\kappa\int_{\xi_0}^{\xi}\frac{{\rm d}\xi_1}{Z(\xi_1, r)}\right] \times \int_{\xi_0}^{\xi}\frac{1 + {\rm i}\varepsilon}{Z(\xi_1, r)}\exp\left\{{\rm i}\left[\alpha\xi_1 - 2\tan^{-1}\xi_1 - \kappa(1 + {\rm i}\varepsilon)\int_{\xi_0}^{\xi_1}\frac{{\rm d}\xi_2}{Z(\xi_2, r)}\right]\right\}\frac{{\rm d}\xi_1}{1 + \xi_1^2}, \quad (7)$$

where $\kappa = bu_3\gamma/2$; $\varepsilon = (\delta T_2)^{-1}$; $Z(\xi, r) = [(1 + \Omega_{\rm d})^2 + v|E_1|^6]^{1/2}$; $a = b\Delta k'/2$; $u_1 = q_3\eta_0/(8\hbar\delta)$; $u_3 = \mu_{41}\eta_0(2\hbar\delta)^{-1}$; $v = 4u_1^2$; $\Omega_{\rm d} = \Omega_2/\delta = \Omega_{\rm p}/|E_1|^2$; $\gamma = 4\pi\omega_3^2\mu_{14}N/(k_3')$; $\Delta k' = 3k_1 - k_3'$ is the frequency detuning; ω_2 and k_3 are the frequency and the wave number of harmonic radiation; and N is the density of a medium. Expression (7) is given within a constant phase factor.

The analysis of (7) shows that efficient third-harmonic generation in media with positive dispersion under condition of strong focusing of laser radiation is possible and can be caused by the influence of both the optical Stark effect and saturation. The latter is similar to the results of Ref. [12], where the third-harmonic generation under two-photon resonance conditions was analysed. The harmonic generation is caused by a change in conversion conditions up- and downstream the beam waist, which violates the conditions of complete radiation transfer from the harmonic generated upstream from the beam waist to the fundamental radiation downstream from the waist.

Fig. 1 presents the dependence of the harmonic intensity on the fundamental-radiation intensity for $\kappa = 1$, $\Omega_{\rm p} = 10^{-12}$ cm^2 W^{-1}, $\alpha = 0$, and $\varepsilon = 0$. At low fundamental-radiation intensities, the harmonic signal is caused by an inexact fulfilment of tight-focusing conditions (in the calculations, $L/b = 40$). As the pump intensity is increased, the harmonic intensity sharply increases, and the dependence becomes close to the fifth-order power dependence. As the pump intensity is increased further, the harmonic intensity saturates. In the experiments, the power dependence of the harmonic signal with a power of 4.2 was observed [8].

The optimum pressures obtained from expression (7) for three lenses with different focal distances for the parameters

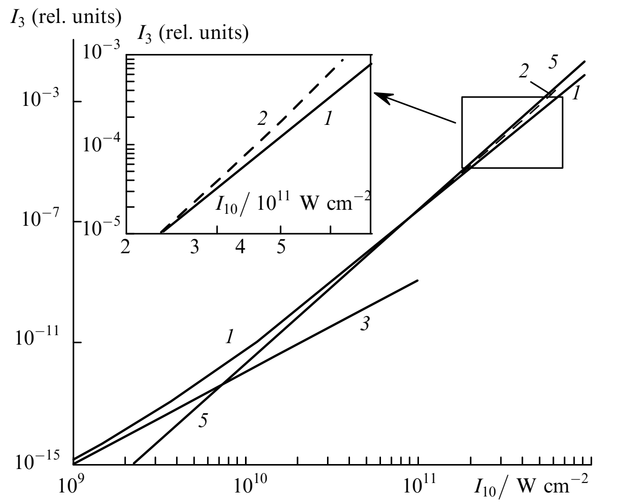

Figure 1. Dependence of the third-harmonic intensity I_3 on the fundamental-radiation intensity I_{10} for the governing influence of the optical Stark effect (1) or Kerr nonlinearities (the dashed curve 2). The straight lines correspond to the power dependences of third (3) and fifth (5) orders. In the insert, the power dependence 5 is omitted.

close to the experimental parameters used in Ref. [8] are in the ratio 0.5:1:1.2, and the corresponding experimental values were in the ratio 0.8:1:1.1. (Note that the change in the optimum pressure is also caused by the resonance absorption of the harmonic signal.) In this case, the harmonic signals should be in the ratio 0.15:1:0.72, whereas the corresponding experimental values were approximately in the ratio 0.125: :1:0.73. Thus, we obtained a good agreement with the experiment, which suggests that the harmonic generation in this case is determined by the optical Stark effect.

Far from resonance, the violation of conditions for compensation and harmonic generation under tight-focusing conditions can be described in terms of Kerr nonlinearities [13 – 16]. The dashed line 2 in the figure shows the dependence calculated by the method [16] for $\alpha = -1$ and $\lambda = (-10^{-12}$ W cm$^{-2})/I_{10}$. (The parameter λ describes the effect of the nonlinearity $\chi^{(3)}(\omega_3; \omega_3, \omega, -\omega)$ [16].) In this case, as one can see from the figure, the influence of optical Kerr and Stark effects at relatively low fundamental-radiation intensities leads to identical results. A nearly fifth-order power dependence of the third-harmonic signal on the fundamental radiation intensity was observed in the experiments [17 – 19], where the third-harmonic generation in rare gases was studied. To differentiate between harmonic generation mecha-nisms, one may take the dependence on the density of a medium as a criterion. At small densities, the influence of Kerr effect leads to the fourth-order power dependence, and in the case where the harmonic generation is caused by the six-wave process, the dependence should be quadratic [10]. In the expe-riment [18], the dependence was close to the fourth-order dependence.

Thus, the Stark shift and the resonance parts of Kerr nonlinearites are caused by the same change in the state of a medium, and their effects cannot be separated. The choice of a way for describing these effects depends on a specific experimental situation. If it is required to take into account the Stark shift, one should exclude from the analysis the resonance contribution of the corresponding terms in all types of nonlinearities of different orders.

References

1. Sobelman I I Atomic Spectra and Radiative Transitions (Berlin: Springer-Verlag, 1979)
2. Delone N B, Krainov V P Osnovy Nelineinoi Optiki Atomarnykh Gazov (Foundations of Nonlinear Optics of Atomic Gases) (Moscow: Nauka, 1986)
3. Bloembergen N Nonlinear Optics (New York: Benjamin, 1965)
4. Butylkin V S, Kaplan A E, Khronopullo Yu G, Yakubovich E I Rezonansnye Vzaimodeistviya Sveta s Veshchestvom (Resonance Interactions of Light with Matter) (Moscow: Nauka, 1977)
5. Klyshko D N Fizicheskie Osnovy Kvantovoi Elektroniki (Physical Foundations of Quantum Electronics) (Moscow: Nauka, 1986)
6. Reintjes J F Nonlinear Optical Parametric Processes in Liquids and Gases (New York: Academic Press, 1984)
7. Mittleman D M, Douglass D C, Henis Z, Wood II O R, Freeman R R, McIlrath T J J. Opt. Soc. Am. B **13** 170 (1996)
8. Kutzner J, Zacharias H Appl. Phys. B **66** 571 (1998)
9. Puell H, Vidal C R Phys. Rev. A **14** 2225 (1976)
10. Dubovik A N Vestn. Mosk. Univ., Ser. 3 **18** 82 (1977)
11. Lago A, Hilber G, Wallenstein R Phys. Rev. A **36** 3827 (1987)
12. Anikin V I, Gora V D, Drabovich K N, Dubovik A N Kvantovaya Elektron. (Moscow) **3** 330 (1976) [Sov. J. Quantum Electron. **6** 174 (1976)]
13. Drabovich K N, Kulagin I A, Usmanov T Kvantovaya Elektron. (Moscow) **12** 616 (1985) [Sov. J. Quantum Electron. **15** 402 (1985)]
14. Butylkin V S, Fedorova M B Kvantovaya Elektron. (Moscow) **21** 158 (1994) [Quantum Electron. **24** 148 (1994)]
15. Butylkin V S Kvantovaya Elektron. (Moscow) **25** 541 (1998) [Quantum Electron. **28** 526 (1998)]
16. Kulagin I A, Usmanov T Kvantovaya Elektron. (Moscow) **25** 1121 (1998) [Quantum Electron. **28** 1089 (1998)]
17. Reintjes J, Dlabal M, Tankersley L L Proceedings of the Conference on Laser Techniques for Extreme Ultraviolet Spectroscopy, Boulder, CO, 1982 (Washington, DC: Optical Society of America, 1983) p. 491
18. Ganeev R A, Gorbushin V V, Kulagin I A, Usmanov T Appl. Phys. B **41** 69 (1986)
19. L'Hullier A, Lompre L A, Ferray M, Mainfray G Europhys. Lett. **5** 601 (1988)