# Elementary nonlinear optical phenomena

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A new line of research has taken form in recent years that lies at the boundary between nonlinear optics, atomic physics, quantum radiophysics, and plasma physics; this is the study of elementary nonlinear optical phenomena. Such phenomena (which take a high-intensity light field to observe) as the creation of electron pairs, the stimulated Compton effect, many-photon ionization and many-photon excitation of atoms, resonance dissociation of molecules, and the nonlinear surface photoeffect are not only of independent interest. On the one hand, study of these phenomena furthers the development of quantum electrodynamics and quantum mechanics. On the other hand, these phenomena are the basis of a number of astrophysical processes that occur when powerful laser radiation interacts with matter, of resonance chemistry, and of a number of new methods of plasma diagnostics.

The progress in development of lasers now permits us to get field intensities approaching atomic values over a broad range of wavelengths (from several hundred Ångströms to ten microns). Utilization of dye lasers has furnished experimenters with a source of intense light with smoothly variable frequency. Extremely high radiation intensities are attainable at a number of wavelengths. One can achieve the (currently) record-breaking field intensity of up to $10^{11}$ V/cm at the wavelength 1.06 $\mu$m (neodymium glass laser). The considerable volumes of space over which one can obtain strong fields permit one to study not only elementary processes but also cumulative effects.

Elementary nonlinear optical phenomena are occupying an ever greater position in the programs of a number of the major periodically-held conferences (the International Conference on Quantum Electronics, the International Conference on Phenomena in Ionized Gases, the All-Union Conference on Nonlinear Optics). However, it has not been the main topic in any of them. Hence, the idea of organizing an international conference devoted to elementary nonlinear optical phenomena has been actively supported in a number of countries including the USSR. The conference was organized by the Hungarian Academy of Sciences, the L. Eötvös Hungarian Physical Society, and the Central Institute of Physical Studies of the Hungarian Academy of Sciences.

The most impressive section of the conference, both in the number of participants and in the number of presented papers, was that on nonlinear photoionization of atoms and molecules. We should note that studies of nonlinear effects arising in molecules were not represented widely enough at the conference. As for atoms, we can distinguish three fundamental problems that were discussed in detail at the conference.

The first problem is how the nature of the process of many-photon ionization of an atom depends on the concrete spectrum of the bound states of the electron and the radiation frequency. Studies performed in recent years at the Institute of Physics of the Academy of Sciences have shown that one can distinguish two typical cases: the direct and resonance ionization processes.[1] It is called a direct process of many-photon ionization whenever the electron goes while absorbing field quanta from the ground state to the continuum via a series of virtual states whose energies $k\hbar\omega$ differ from the energy $E_i(\mathscr{E})$ of the bound states of the electron perturbed by the radiation field. For the direct process, the probability W of ionization is related to the field intensity $\mathscr{E}$ by the relation $W = \alpha_{k_0}\mathscr{E}^{2k_0}$, where $k_0$ is the number of quanta absorbed in ionization, $k_0 = \langle (I/\hbar\omega) + 1\rangle$, and $\alpha_{k_0}$ is the cross-section of the process. Comparison of the experimental data on the cross-sections of two-, three-, four-, and five-photon ionization processes, which were mainly measured at the Institute of Physics of the Academy of Sciences, with the calculations from perturbation theory in the order corresponding to $k_0$ showed that the first non-vanishing order of perturbation theory describes well the experimental data quantitatively.[2]

The process of many-photon ionization is termed a resonance process whenever the electron arrives in a state that proves to be real as field quanta are absorbed, and the relationship $k\hbar\omega = E_i(\mathscr{E})$ is obeyed. The possibility of appearance of many-photon resonance, in contrast to single-photon resonance, is governed by two parameters: not only the frequency, but also the field intensity of the radiation. Resonance can be induced by a strong field that perturbs the bound states and substantially alters their energies and widths. When resonance exists, the ionization process occurs in two stages: many-photon resonance excitation of the atom, and ionization of the excited atom. The appearance of an intermediate many-photon resonance substantially changes the nature of the process of many-photon ionization of an atom: it increases the probability of ionization and alters the functional relationship of the probability to the radiation intensity. Depending on the relationship between the resonance mismatch, the broadening, and the change in the energy of the resonance state perturbed by the radiation field, one can observe a $W(\mathscr{E})$ relationship that is either smoother or steeper than a power function with exponent $2k_0$.

The process of resonance many-photon ionization has proved interesting not only per se. Observation of a resonance process has made it possible to study the perturbation of atomic levels in a strong light field. The classical methods, which involve using an auxiliary light to observe the excited state, as has been done by the group of Bonch-Bruevich (USSR)[3] for resonance perturbation and by P. Platz (France)[4] for non-resonance perturbation cannot be used in the general case. Actually, the probability of ionization of the excited state by action of the strong main field is very large. Thus the lifetime in the excited state is very short: it is determined not by the spontaneous downward transition, but by the stimulated upward transition. Yet, even the first studies of the resonance ionization process performed in the Institute of Physics of the Academy of Sciences[1] showed that, if one observes changes in the $W = f(\mathscr{E})$ relationship involving a change in the frequency and intensity of the radiation, one can measure both the energy of the perturbed resonance state and its width, i.e., get full information on the perturbation. This method has been used in a series of studies conducted in the Central Institute of Physical Studies of the Academy of Sciences of Hungary by the group of J. Bakos.[5] Thus, it has been possible to measure the change in the energy of the transition from the $2^3$S state in the helium atom to states of principal quantum numbers 11—15 at field intensities of the order of $10^6$ V/cm. Data on perturbation of high levels in a strong field are of great interest. Comparison of such data with calculations of the dynamic polarizability of atoms performed by perturbation theory permits us, in particular, to establish the limit of applicability of the first non-vanishing order. In this regard, it is interesting to note the calculations of the dynamic polarizability of the hydrogen atom in the first excited states which were carried out with account taken of the terms proportional to $\mathscr{E}^2$, $\mathscr{E}^4$, and $\mathscr{E}^6$ (L. Gontie and M. Trahin, France (Saclay)).[6] It turned out that the higher-order terms give a contribution comparable with that of the term $\sim \mathscr{E}^2$, even at $\mathscr{E} \sim 10^7$ V/cm.

Finally, the third problem that was widely discussed at the conference was that of the spectrum of quasi-steady states of an atom-field system in which the perturbation cannot be considered to be small. Various experimental and theoretical data that can be adduced to analyze the spectrum give contradictory results. Thus, one possibility consists in extrapolating to a strong field the experimental and theoretical data that describe the perturbation in a relatively weak field. If one estimates the ionization broadening of the bound states in a strong field from the known cross-sections for one-photon ionization and the recently-measured cross-sections for many-photon ionization[1] (including that from excited states), then it turns out that practically the entire spectrum of bound states must merge into a continuum, even at a field intensity of several times $10^7$ V/cm. Appearance of a continuum is also predicted by extrapolation of data on the dynamic polarization of atoms as obtained in the first non-vanishing approximation of perturbation theory.[2] Application of the quasienergy method has not yet given any general results for a real atom.[7] We should assume that the quasienergy spectrum of an atom in a strong light field should also merge into a continuum, owing to the high probability of ionization from the excited states. Yet, studies of the frequency-dependence of the process of many-photon ionization of noble-gas atoms at a field intensity of $\sim 5 \times 10^7$ V/cm have revealed a resonance alteration in the nature of the process. This indicates that the spectrum of bound states is discrete.[8] Currently theory is lagging behind experiment in the study of this problem, which is of a fundamental nature. Thus, e.g., it is even difficult to make a qualitative analysis of the experimental data. It is now hard to point out the optimal method for solving the problem of the spectrum of quasi-steady states of the atom-field system. However, we should note that the quasienergy method[7] has made it possible to get important results in a number of analogous problems, including prediction of the qualitative variation of the light-scattering spectrum.[9]

In the section devoted to nonlinear photoemission from solids, they mainly discussed papers given by the hosts of the conference, who were researchers from the Central Institute of Physical Studies of the Academy of Sciences of Hungary. Although the nonlinear surface photoeffect has attracted the attention of large groups of physicists in various countries, only the group of G. Farkas in this institute has yet obtained experimental results. The methododological success of this group has involved using ultrashort laser pulses. When the surface of a metal or dielectric is illuminated with radiation of a mode-synchronized laser (with a pulse duration of $\sim 10^{-11}$ sec), the target cannot heat up strongly. Thus we can neglect the electrons formed by the Richardson effect as compared by those formed by the non-linear photoeffect. Experimental study of the surface photoeffect has thus far permitted elucidating two important facts.[10] First, a number of metals and dielectrics exhibit a nonlinear emission of electrons when irradiated with radiation of wavelengths 0.67 and 1.06 $\mu$m. The latter is well described by the power law $J = \alpha_{k_0}\mathscr{E}^{2k_0}$, where $\alpha_{k_0}$ is the cross-section of the process, $k_0 = \langle (A/\hbar\omega) + 1 \rangle$, and A is the work function. The experimental data obtained for the degree of nonlinearity over the range $2 \leq k_0 \leq 5$ permit one to determine the cross-section $\alpha_{k_0}$ of the process and to compare it with the results of calculations that take account of the details of the process. Second, when a gold target was irradiated, a deviation from the power law with exponent $k_0 = 4$ was found as the intensity of radiation at $\lambda = 1.06$ $\mu$m was increased. Control experiments performed at the same intensity, but at $\lambda = 0.53$ $\mu$m, showed no deviation from the power law. In line with the predictions of the theory, the effect of deviation from a power law can be caused by the tunneling nature of the photoemission process. To use the surface photoeffect for studying the transition from a many-photon type of ionization process to a tunneling type is perhaps currently the only possibility of experimentally testing the generally-known theory of ionization in a light field originated by L. V. Keldysh. We should note that development of the theory of the nonlinear surface photoeffect as yet lags behind the experimental progress. Yet studies of the nonlinear surface photoeffect can give important information on the structure of the potential at the solid-vacuum boundary, on the depth of penetration of the strong field into the metal or dielectric, and also on the statistics of the laser radiation.

The overwhelming majority of the papers discussed in the section on interaction of a free electron with intense laser radiation dealt with the theory of various elementary effects. Individual special cases of the process of pair creation in a vacuum by the action of a powerful light field were broadly discussed. As we know, studies of such processes are of great interest for quantum electrodynamics. In a vacuum, the process of creation of electron pairs becomes effective when the condition $(E/mc^2)\mathscr{E}/\mathscr{E}_{cr} \sim 1$ is fulfilled, where E is the energy of the electron, $\mathscr{E}$ is the field intensity, and $\mathscr{E}_{cr} = m^2c^3/e\hbar$. A field intensity $\mathscr{E} \sim 10^{15}$ V/cm is needed to satisfy this condition. This is 4–5 orders of magnitude higher than the record-breaking achievements of modern laser technology. However, in the field of a nucleus, where the process of pair creation occurs during scattering, it becomes effective at a considerably lower light field intensity.[11] For this to happen, it suffices that a non-relativistic electron can on the average become relativistic within the period of the wave. As was stated above, the needed field intensity $\mathscr{E} \sim 10^{11}$ V/cm is attainable at the limit of modern potentialities. This makes it pertinent to perform detailed theoretical studies of the process of pair creation in the field of a nucleus, since we should expect the appearance of experimental data in the very near future.

The nature of the Compton effect changes qualitatively at high field intensities: the same two-photon scattering process no longer is spontaneous in nature, but stimulated. Since the efficiency of stimulated scattering increases with increasing radiation intensity, the Compton effect plays a substantial role in the interaction of laser radiation with a gas or plasma, even at the easily attainable field intensity $\mathscr{E} \sim 10^6$ V/cm. Energy can be transferred to an electron only when the frequency and angular spectra of the radiation have finite widths. Since the number of photons is conserved in the stimulated (two-photon) Compton effect, the heating of the electrons is accompanied by a red shift in the radiation spectrum, as is observed experimentally.[12] The large intensity of the monochromatic field of laser radiation also permits one to observe the Kapitza-Dirac effect. As we know, the latter is a special case of two-photon stimulated Compton scattering, or elastic scattering.[13] Interestingly, the stimulated Compton effect can be described in the language of classical physics[14] if the time of passage of the electron through a distance of the order of the wavelength is much greater than the period of the field. The heating of electrons here can be described as resulting from Brownian movement in the field of the gradient force, while the scattering of electrons results from motion of the electron in the rapidly varying field.

The Schwarz-Hora effect was discussed[19] anew at the conference. As we know, this effect consists in emission from a screen acted on by an electron beam modulated by laser radiation. The modulation was performed by illuminating a thin film through which the beam was passing. Emission from the screen was observed at a frequency that corresponded to the modulation frequency. The interpretation of these first[15] and as yet only results has led to development of two new lines of research: the quantum theory of modulation of an electron beam by light[16] and the theory of coherent transition radiation.[16,17] From the standpoint of quantum theory, after an electron has passed through a medium lying in a light field, it does not have a definite energy and momentum, and its wave function is a superposition of states that arise from stimulated absorption and emission of n quanta of light. The interference of these states modulates the density of the beam in space. When the modulated flux of electrons interacts with the surface of the screen, coherent transition luminescence arises in addition to the continuous emission spectrum (transition radiation, bremsstrahlung, luminescence).[17] The intensity of the coherent radiation, which has the frequency of the modulation of the electron beam, is proportional to the square of the current.

Thus, on the one hand, it has been possible to treat theoretically the physical phenomena that lead to modulation of the electron beam and to appearance of the coherent transition radiation. On the other hand, a detailed quantitative analysis performed by B. Ya. Zel'dovich has shown that under the experimental conditions of[15] the maximum possible intensity of coherent radiation is $10^4$ times smaller than the amount reported in[15], while the radiation having a continuous spectrum has an intensity, $10^5$ times greater than the coherent radiation. Evidently, the experiment[15] must be repeated, and a series of experiments must be performed with variation of the fundamental parameters, as well as a careful quantitative analysis of the experimental data, so as to permit the first conclusions on the modulation of an electron beam by light and on coherent transition radiation.

Besides the above-mentioned lack of attention to processes occurring in molecules, a substantial defect of the conference was that just as little attention was paid to processes of many-photon light scattering by atoms. The paper by S. Kilich (Poland) gave only a general picture of the entire variety of different nonlinear effects in light scattering.[20]

A number of the participants gave a general estimate of the conference in the closing session. A. M. Bonch-Bruevich (USSR), S. Kilich (Poland), and H. R. Reiss (USA) noted the great importance of the conference for development of studies in the field of elementary nonlinear optical phenomena, the good preparation of the conference (chairman of the Organizing Committee J. Bakos, Hungary), and the excellent organization of the conference (the Division of Physical Optics of the Central Institute of Physical Studies of the Academy of Sciences of Hungary). The opinion was universal that conferences must be held periodically on this topic. On the one hand, it would be desirable here to expand the discussion of a number of problems (interaction of intense light with molecules, many-photon Raman phenomena, the stimulated Compton effect, and the stimulated inverse bremsstrahlung effect), and on the other hand, to restrict the topic more strictly to elementary processes proper, without treating cumulative or averaged effects that arise when intense light interacts with macroscopic objects.

It was decided at the closing session of the Organizing Committee to publish the review papers. The Central Institute of Physical Studies of Hungary assumed this responsibility (editor J. Bakos). The collected volume of papers has now been published. It was decided to hold the next conference in 1975 in Hungary. The chairman of the Organizing Committee continues to be J. Bakos (Central Institute of Physical Studies of the Academy of Sciences of the Hungarian People's Republic, POB-49, Budapest 114, Hungary).

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[1] G. Delone, Invited Papers of the Conference on the Interaction of Strong Electromagnetic Fields with Electrons, Budapest, 1973.

[2] L. Rapoport, ibid., p. 99.

[3] A. Bonch-Bruevich, ibid., p. 255.

[4] R. Rapoular and P. Platz, ibid., p. 211.

[5] J. Bakos, A. Kiss, L. Szabo, and M. Tendler, Reprint KFKI 69, 1972.

[6] I. Gontie and M. Trahin, Reprint CEA (Saclay), No. 112, 1972.

[7] Ya. B. Zel'dovich, see Ref. 1, p. 5; Usp. Fiz. Nauk 110, 139 (1973) [Sov. Phys.-Uspekhi 17, 427 (1973)].

[8] D. T. Alimov, N. K. Berezhetskaya, G. A. Delone, and N. B. Delone, Kr. Soobshch. Fiz. (FIAN SSSR), No. 11, 21 (1971); Zh. Eksp. Teor. Fiz. **64**, 1178 (1973) [Sov. Phys.-JETP **37**, 599 (1973)]. G. Baravian, R. Benattar, J. Bretagne, G. Callede, J. L. Godart, and G. Sultan, Appl. Phys. Lett. **18**, 387 (1971).

[9] V. Kovarsky, see Ref. 1, p. 125.

[10] G. Farkas, see Ref. 1, p. 179.

[11] E. Bunkin, see Ref. 1, p. 59.

[12] I. K. Krasyuk, P. P. Pashinin, and A. M. Prokhorov, ZhÉTF Pis. Red. **12**, 439 (1970) [JETP Lett. **12**, 305 (1970)]; M. Decroisette, G. Piar, and F. Floux, Phys. Lett. **A32**, 249 (1970).

[13] L. S. Bartell, H. B. Thompson, and R. R. Roskos, Phys. Rev. Lett. **14**, 851 (1965); H. Schwartz, H. A. Tourtellotte, and W. W. Gaertner, Phys. Lett. **19**, 202 (1965).

[14] B. Ya. Zel'dovich, Annotation of papers given at the conference (Ref. 1), p. 27.

[15] H. Schwarz and H. Hora, Appl. Phys. Lett. **15**, 349 (1969).

[16] D. A. Varshalovich and M. A. D'yakonov, ZhÉTF Pis. Red. **11**, 594 (1970) [JETP Lett. **11**, 411 (1970); Zh. Éksp. Teor. Fiz. **60**, 90 (1971) [Sov. Phys.-JETP **33**, 51 (1971)].

[17] P. L. Rubin, ZhÉTF Pis. Red. **11**, 356 (1970) [JETP Lett. **11**, 239 (1970)].

[18] B. Ya. Zel'dovich, Zh. Éksp. Teor. Fiz. **61**, 135 (1971) [Sov. Phys.-JETP **34**, 70 (1972)].

[19] H. Schwarz, see Ref. 1, p. 39.

[20] S. Kilich, see Ref. 1, p. 279.