Presentations

Nonlinear Optics Division, Institute of Physics, Adam Mickiewicz University, Poznań, Poland

Research in nonlinear molecular optics began in Poznań in the Laboratory of the Optics of Dielectrics of the Polish Academy of Sciences (1955–1964), and was continued in the Chair of Molecular Physics (1965–1969) and the Institute of Physics of Adam Mickiewicz University (1969–1972).

In its present form, the Division of Nonlinear Optics was organized in 1972; since then it has been headed and directed by Professor Dr. Stanisław Kielich. At the time of its foundation, its staff comprised 1 professor, 1 senior lecturer, 1 doctor, 10 assistants, 2 postgraduate students, and 2 technicians (althogether 17 persons).

The research work carried out in 1958–1972 was discussed in the previous review, Optica Applicata, Vol. 4 (1974), 39. The essential results in 1972–1980 are given in *European Who's Who in Quantum Electronics* (edited by F. P. Schäfer, Göttingen, Druckerei C. Krummrat).

The research programme was concentrated on:

a) linear and nonlinear molecular light scattering in dense molecular systems, and

b) nonlinear electro-magnetic and optical phenomena in isotropic media and solids.

Present data

Head of division: Prof. Dr. S. Kielich,
Senior Staff: Senior Lecturer K. Flatau, M. Sc.,
Drs: M. Kozierowski, Z. Ożgo, W. Alexiewicz, T. Bancewicz, J. Buchert, M. Kotłowska, A. Planner, L. Sczaniecki, J. Skupiński, P. Szlachetka, R. Tanaś, L. Wołejko, S. Woźniak, R. Zawodny.

Total personnel 31, comprising 5 technicians.

Research programme

A. Theory of multi-photon resonant and non-resonant Rayleigh and Raman spectroscopy.

B. Molecular dynamics of nonlinear optical and electro-magnetic phenomena in gases and dense systems.

C. Photon statistics, squeezed states of electromagnetic field and cooperativity in nonlinear optical phenomena.

D. Theory of multi-photon lasers and experimental investigations.

Book and review articles

1. Monograph on *Nonlinear Molecular Optics* by S. KIELICH (paper 1), Russian amplified translation.

2. Comprehensive monographic article on the most recent achievements in multiphoton molecular spectroscopy, dealing essentially with the results obtained in our division, i.a., in the fields of the spectral theory of Rayleigh light scattering, the theory of multi-

photon scattering processes of the Raman type, and the role of the polarization state of the incident beam (papers: 2, 62–65, 68).

3. Review articles (papers: 3, 4, 14–17, 34, 43) on photon anticorrelation, presented simultaneously in the form of a sectional report at the International Conference in Quantum Electronics and Nonlinear Optics (EKON–80) in Poznań, giving the earlier results obtained in our Division in the field of photon statistics which led to the discovery of this nonlinear quantal effect for processes of harmonics generation of laser light, nonlinear polarizational effects, and hyper-Raman scatterings.

4. Publications (papers: 5, 35–40, 43–45) discussing the possible occurrence of squeezed states of the electromagnetic field having, like photon antibunching, no classical counterpart, thus providing a test of the correctness of QED. These tests were carried out for resonance fluorescence of two interacting atoms, nonlinear polarizational effects, and the generation of light harmonics. These studies were reviewed at the Conference held in Rydzyna (EOC'83).

5. Experimental and theoretical achievements in the domain of nonlinear electro-optical and magneto-optical phenomena in molecular gases and liquids, reviewed in paper 21.

Theory of multi-photon resonant and non-resonant Rayleigh and Raman spectroscopy

1. A theory of light scattering on correlated molecules has been proposed, taking into account fluctuation of the multipolar molecular fields as well as interference effects (papers: 6, 7, 19, 20, 30–33).

2. Using the methods of spherical algebra and applying the dipole-induced dipole model, a general expression for the effective hyperpolarizability is derived with accuracy to within the second-order of statistical perturbation theory. The results have permitted the application of a computer simulated binary distribution function to calculate the hyper-Rayleigh scattered intensity (paper 33).

3. To describe finite laser bandwidth effects in n-photon processes the phase diffusion model is applied. The n^2-dependence of the linewidth of fluorescent light is proved in the case of n-photon resonance (paper 18).

4. Treating the fluctuations in number ΔN as complex stochastic processes, the problem of higher order time-dependent correlation functions of N is discussed (paper 26).

5. For atoms with closed electron shells, the Van der Waals interactions have been described in terms of orbital form factors and applied to calculate the Ne-Ne potential (paper 42).

6. Experimental temperature and pressure Raman studies on molecular motions in liquid CH_2Cl_2 (paper 67) have provided important information about the reorientational relaxation and coupling mechanism in the liquid.

7. A detailed discussion of symmetric and unsymmetric scattering is carried out for linear, circular and natural polarization of the incident light. Such parameters as the ellipticity, circularity degree, and reversal and depolarization ratios are studied (papers 70, 72).

Molecular dynamics of nonlinear optical and electro-magnetic phenomena in gases and dense systems

1. Expressions for the cross-sections for dipole-magnetic and quadrupole-electric transitions have been obtained and their values for selected quantum atomic transitions calculated. The angular distribution for light scatterings is presented (paper 10).

2. It has been shown that a d.c. magnetic field, acting on a liquid along the propagation direction of circularly polarized light, causes a magneto-dispersional-spatial variation of the refractive index (papers 28, 32, 41).

3. A theory of the nonlinear Faraday effect in atomic systems has been proposed, and the variations in Verdet constant have been calculated applying the Green function method (paper 9).

4. The theory of optical activity has been generalized to include the effect of molecular interactions arising from the induced moments. Thus, the electronic quadrupole terms do not vanish upon statistical averaging (paper 60).

Photon statistics, squeezed states of electromagnetic field and cooperativity in nonlinear optical phenomena

1. The exact analytical solution for propagation of the quantized electromagnetic field in a nonlinear optically isotropic medium has been obtained. It has been shown that the field can emerge from the medium in a squeezed state, the latter being produced by the field itself (self-squeezing)–papers 44, 45.

2. Analytical solutions for the two-atom resonance fluorescence spectrum of symmetric and antisymmetric modes have been obtained without decoupling approximation. It has been moreover shown that the statistics of the scattered light depends on the interatomic separation r_{12} as well as on the direction of observation. If the resonant field is strong and the cooperative damping taken into account, the spectrum consists of seven lines and the initial value of the intensity correlation function is independent of dipole-dipole interaction (papers: 24, 25, 34).

3. The time-dependent spectrum of resonance fluorescence from two-level atoms is obtained after a strong resonant exciting laser field is turned off abruptly. The speed of *undressing* of the atomic states is considered (paper 29).

4. It has been shown that, for great intensities of the driving field, the absorption spectrum is negative in a certain range of frequencies, and that the form of this absorption spectrum is considerably modified by dipole-dipole interaction between the atoms and a new region of negative absorption can appear (paper 58).

5. Contrary to the steady-state régime, the transient régime of resonance fluorescence from a two-level atom admits of squeezing for arbitrary intensity of the exciting field (papers 59, 71).

6. The equivalence of quantum optical phenomena, described in terms of Langevin and Fokker-Planck equations, is proved. The role of multiplicative stochastic processes in quantum optics is discussed. A very simple way of calculating the diffusion coefficients is proposed (paper 57).

7. Squeezing in a dynamical stochastic system governed by an operator Langevin equation with multiplicative Ornstein-Uhlenbeck noise is presented. The Langevin equation is solved by the small parameter method. The solution proposed implies no boson anti-bunching (paper 61).

Theory of multi-photon laser and experimental investigations

1. A new operator form of perturbation calculus leading to effective Hamiltonians of interaction between the system and the electromagnetic field has been proposed. Moreover, generalized Maxwell-Bloch equations have been derived (paper 46).

2. A quantum-mechanical theory of the multi-photon laser that has been elaborated proved the existence of bistability and non-equilibrium phase transitions of the 1st kind (papers 12, 13, 27).

3. Suspensions of solid absorbing particles, arising when a liquid is damaged by a focussed laser beam, are shown to increase or decrease feedback in pulse lasers (paper 11).

4. The construction and operation of a passively mode-locked ruby laser and its application to the excitation of transient stimulated Raman scattering (TSRS) is reported (papers 22, 23).

5. An experimental study of laser beam defocussing has been carried out for the case when the medium is subjected to heating for time comparable with that of its thermal relaxation. This provides a method for the direct determination of the heat diffusivity coefficient (papers 8, 61).

6. Patent No. 108369, adjudicated by the Bureau of Patents of the People's Republic of Poland, has been obtained for an *Instrument for measuring the thickness of transparent objects* (paper 66).

7. Papers 47–52 are devoted to measurements of the kinetics of the first intermediate of visual pigments (bovine rhodopsin, isorhodopsin, squid rhodopsin) as well as the kinetics of energy transfer between different pigments by using picosecond absorption spectroscopy. The effect of excited annihilation (described in paper 47) provides an important channel for radiation-less decay which is competitive with radiative deexcitation.

8. The effect of hot carrier relaxation in semiconductors was studied in paper 50. Photogenerated carrier densities and excited intensities are so high as to create non-equilibrium phonon populations making it possible to study some of the fundamental energy decay routes.

9. Investigations in the field of new solid state, tunable laser materials are described (papers 53–55). To compute high power levels of the line shape saturation, nonradiative loss processes, self mode-locking and *hole burning* in vibronic lasers the properties of emerald, associated with vibronic terminated transitions and the temporal bahaviour of an optical phonon are required. These lasers are potentially important in technology, as the basic element of very broad-band optical amplifiers and tunable oscillators (in emerald 700–850 nm). Emerald, as a solid state material, may be Q-switched or mode-locked by an active method or by using dyes as saturable absorbers, and can generate pico- or femtosecond light pulses.

10. Paper 56 deals with the properties of a neodymium glass mode-locked laser in nonconventional colliding pulse of an anti-resonant arrangement, in which the obtained picosecond pulses were about 3 times shorter than in traditional desing.

Cooperation with other groups

Our division has published a number of papers in cooperation with foreign laboratories: Centre de Recherche C.N.R.S. Paul Pascal Talence, France (papers 8, 61), Voronezh State University, USSR (papers 9, 10), Université d'Angers, France (papers 11, 22, 23), Palacký University, Olomouc, Czechoslovakia (papers 14–17), University of Rochester, USA (paper 29), Stony Brook University and Pennsylvania State University, USA (papers 19, 33, 73), the Florida State University, USA (papers 41, 42, 60), and the City College of New York, USA (papers 47–56).

Measurement facilities available in our Division

An experimental group of four is active in our division, though our research is predominantly theoretical.

1. Recording laser-induced Raman spectra: measurement range of Raman shifts 0–4000 cm^{-1}, maximum resolving power 1 cm^{-1}, form of sample – liquid, powder, single crystal.

2. Recording in infrared spectra: range of transmission measurements 400–4000 cm^{-1}, resolving power 1–5 cm^{-1}, form of sample – liquid, powder in disc form, transmission coefficient measurement vs. temperature in the range 0–250°C.

3. Nd: glass mode-locked picosecond laser and detection system. No unique apparatus is available; with regard to the scarcity thereof in Poland, we should maybe include in this group a JRS–S1 Jeol Ltd. (Japan) Raman spectrophotometer, made in 1973.

We intend to continue our research work on the same topics, particularly those in items 1 and 2. Obviously, we shall follow any novel line of research that may emerge due to the rapid progress of physics in our days.

Papers published in 1980-1983

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21. S. KIELICH, *Electro-optic and magneto-optic phenomena in molecular gases and liquids*, [in] *Electro-optic and magneto-optics methods*, Ed. L. Sobczyk, PWN, Warszawa 1983, pp. 9–41.

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