lined in that work, the following protocol can be used in order to realize the quantum teleportation [19]. We imagine a molecular system containing three characteristic cofactors: A, B, and C. At the first stage, A is reduced, and in the system A^-BC, the electron spin in the anion radical A^- is prepared, employing microwave pulses, in a quantum-coherent state in the resonator of a pulsed EPR spectrometer. At the second stage, a light pulse produces the spin-correlated pair B^+C^- of ion radicals, yielding the three-spin system $A^-B^+C^-$. At the third stage, the A^-B^+ pair should recombine. This recombination is a spin-dependent process. As a rule, recombination is allowed for the singlet spin state in the pair A^-B^+. Eventually, we obtain the system ABC^-, in which the spin of the anion radical C^- is in the coherent state that is related to the coherent state of the anion radical A^- by the well-known unitary transformation. According to this protocol, we thus prepare the C^- electron spin in the coherent state by quantum teleportation of the coherence of another anion radical, A^-. To measure the coherent state of C^-, the methods of pulsed EPR spectroscopy can be used. To effect quantum teleportation, a photochemical process can therefore be used to produce the spin-correlated pair B^+C^-, a quantum communication channel, and to use the recombination of A^-B^+ for the projection of this pair on the singlet state.

To realize the above quantum teleportation protocol, there is good reason to use a natural photosynthetic reaction center (RC) or artificial RC models. A pair of separated charges P^+Q^- in the RC is a perfect quantum teleportation channel in a system of electron spins. The problem with this approach consists in the inclusion into the system of the reduced A^-, the carrier of the initial quantum state. Such a modification of the reaction center is basically possible.

5. Conclusion

That the electron spins play an important part in the making of chemical bonds has been well known since the emergence of the Heitler–London theory. For a long time, it was believed that the electron spin states are conserved in an elementary chemical event (Wigner's rule). However, time-resolved EPR experiments showed that short-lived intermediate states — spin-correlated pairs — form in the course of an elementary photochemical or photophysical event. The spin dynamics in these pairs manifest themselves in the unusual properties of the EPR signal. The study of spin polarization in the course of photo-induced processes has come to be an important part of a new scientific area — spin chemistry [20]. It is hoped that spin-dependent photo-induced processes will also find use in quantum computing.

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The role of electron paramagnetic resonance in the development of quantum electronics: facts and comments

A A Manenkov

1. Introduction

In 2004, the scientific community celebrated two milestones, 60 years of electron paramagnetic resonance (EPR) and 50 years of quantum electronics (QE).

Today, we have one more historic moment to celebrate, 60 years of the Zavoisky Kazan Physical-Technical Institute (KFTI). There is in fact a close historical connection between these three anniversaries. E K Zavoisky's 1944 discovery — and the subsequent development — of EPR has fundamentally influenced the making and development of quantum electronics, and the KFTI that bears Zavoisky's name is the leading research institute in the field of EPR. It is this historical connection that served as the stimulus for this paper.

The paper briefly reviews the early development of quantum electronics, including the formulation of its basic principles; the first ammonia-beam quantum oscillator (maser) and the first paramagnetic crystal-based quantum amplifiers (EPR masers); practical EPR maser schemes and their application; and progress from microwave EPR masers to optical quantum oscillators and amplifiers (lasers). The

paper also discusses the feedback of QE on research in the field of EPR.

Finally, some historical facts are presented to show the difficulties in perceiving the EPR and QE ideas at the early stage of their development.

2. The beginning of quantum electronics

Quantum electronics is generally accepted to have begun in 1954, the publication year of the seminal papers [1, 2] that formulated the basic principles underlying the electromagnetic radiation generation due to stimulated emission of atomic systems in quantum transitions and which reported creating the first QE device, a molecular generator (later called *maser*) operating on a beam of ammonia molecules [2].

The 50th anniversary of QE was widely celebrated in Russia and elsewhere — in particular, at the special topical symposium of the International Quantum Electronics Conference (IQEC) and Conference on Lasers and Electrooptics (CLEO), 17 May, 2004, San Francisco, USA, where the following five talks discussed the history and development of QE:

C H Townes, "Early history of quantum electronics";

N F Ramsey, "Early stimulated emission amplification by an inverted nuclear spin population and H-maser";

N Bloembergen, "Historical comments on the pumping of masers and lasers";

A A Manenkov, "Pages from the history of quantum electronics research in the Soviet Union";

K Shimoda, "Original studies of quantum coherence."

These talks were subsequently published in the *Journal of Modern Optics* (Vol. 52, No. 12, August 15, 2005). Unfortunately, this source is very difficult if not impossible to find in a Russian scientific library.

As is known, quantum amplifiers and oscillators fundamentally hinge on the inversion of energy level population with respect to the thermodynamic equilibrium distribution. The first observations of the inversion were made at the nuclear spin levels of ^{7}Li in a crystal of LiF (Pound, Purcell, Ramsey, 1950 – 1951), but they did not lead to the creation of the maser, nor were they recognized as suggesting a new principle for amplifying and generating electromagnetic radiation (for a discussion of these experiments, see Ramsey's San Francisco paper cited above [3]).

The breakthrough came with papers by Basov and Prokhorov [1] and Townes et al. [2], which formulated principles for creating quantum amplifiers and oscillators based on beams of molecules using energy selection in a nonuniform electric field. The first successful realization of these principles used the inversion levels of ammonia molecules [2].

The next key step in the development of QE was to use auxiliary radiation to achieve inversion (Basov and Prokhorov [4], 1955). The idea of the method, as illustrated in Fig. 1 in the case of a three-level system, is that the auxiliary electromagnetic radiation of frequency ν_{aux} induces transitions between the upper and lower levels, producing population inversion between one of these two and an intermediate level, thus creating conditions for the generation at the corresponding transition $1 \rightarrow 2$ at the frequency ν_g.

Although initially proposed for molecular beams, Basov and Prokhorov's method of auxiliary radiation (commonly known as electromagnetic pumping) turned out to be universal and valid for any atomic system.

Figure 1. Schematic of the population inversion using auxiliary radiation (pumping). ν_{aux}, auxiliary radiation frequency; ν_g, signal frequency that provides generation.

In particular, the method was found to be very efficient for inverting the population of the spin levels of paramagnetic ions in crystals, leading to the creation of quantum amplifiers known as EPR masers (Bloembergen [5], 1956). In the first successful realizations of the EPR maser, in 1957 – 1958, the quantum amplification effect was obtained on transitions in paramagnetic impurity atoms of Gd^{3+} in ethyl sulfate [6], of Cr^{3+} in cyanide [7, 8], and of Cr^{3+} in corundum (ruby) [9].

We note that the very idea of using the spin levels of paramagnetic atoms in a crystal for creating masers benefited from the strides in EPR spectroscopy, as did the successful realization and further development of these devices. As an example, our idea to use ruby as an active maser medium (Manenkov, Prokhorov [10], 1956) was based on EPR studies of this material [11, 12].

In subsequent studies (see monograph [13] for a review), the paramagnetic ion doping technique as a tool to obtain active maser materials was extended to include other crystals (Cr^{3+}, Fe^{3+} in rutile and tungstates, Gd^{3+} in fluorite, etc).

The studies have shown that ruby is the most efficient maser material owing to its unique combination of spectral, relaxation, dielectric, thermal, and mechanical properties. Ruby masers operating at decimeter and centimeter wavelengths have been developed.

Figure 2 above shows one of the early laboratory mockups of a decimeter ruby maser built in 1958 at the Oscillations Laboratory of the Lebedev Physics Institute, RAS.

3. Applications of EPR masers

Masers as amplifiers of electromagnetic radiation have an extremely low intrinsic noise level (noise in a quantum system is produced by spontaneous emission in the system itself and by thermal emission in the feeder lines [13]). Therefore, immediately following their laboratory realization, EPR masers attracted much attention for application as high-sensitivity microwave detectors in a variety of fields. In the 1960s and 1970s, a series of masers developed in a number of research institutions in the USSR found practical applications in radio astronomy and long-range space telecommunications [14] — work that was awarded the State Prize in 1976. EPR masers were instrumental, in particular, in performing unique experiments for space research.

For example, detailed studies of galactic hydrogen emission at 21 cm and the discovery of new emission lines of highly excited hydrogen in the 8 mm wavelength range (Sorochenko et al. [15], 1969) provided valuable data on the distribution, temperature, density, and dynamics of hydrogen in the galaxy.

Very interesting data on the water content of some space objects were obtained by observing 1.35 cm emission from space (Matvienko et al. [16 – 18], 1986) following the discovery by Townes et al. [19] in 1969.

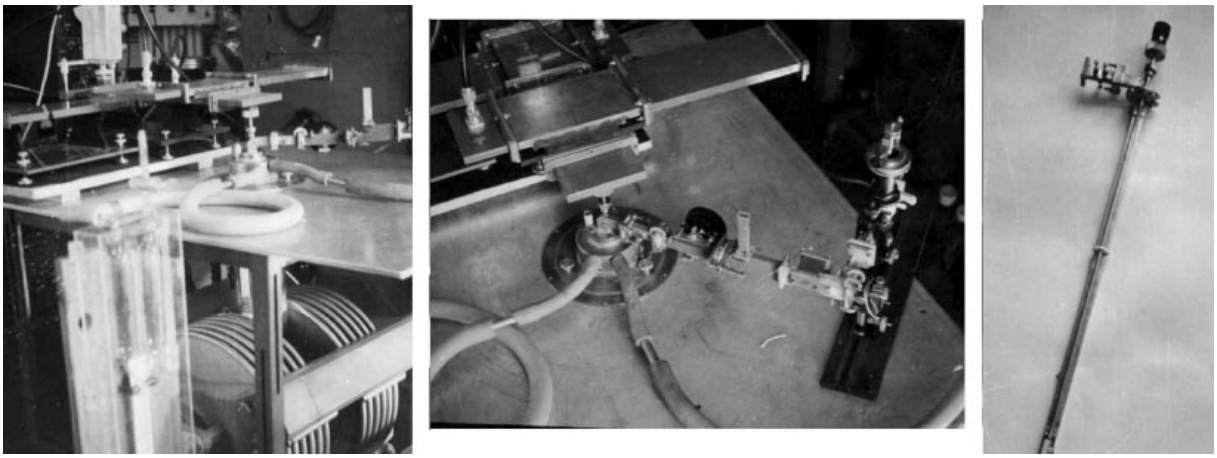

Figure 2. A decimeter EPR maser built in FIAN at 1958. General view (left) and details: center, a signal waveguide at $\lambda_c = 15$ cm and a pumping generator at $\lambda_{pum} = 2.21$ cm; right, two-frequency (signal and pumping) cavity with two feeding waveguides (coaxial for signal, rectangular for pumping).

Finally, planetary radar studies using EPR maser amplifiers have provided new insights into details of what the planets Mercury, Venus, Mars, and Jupiter are like (Kotel'nikov et al. [20], 1962 – 1964).

4. Impact of EPR maser research on the field of EPR

As noted in Section 2, while the EPR maser benefited from progress in EPR spectroscopy, it also provided valuable feedback to the EPR field.

For example, feasibility studies on new crystals for use as active maser media for various wavelength ranges encouraged the development of the EPR spectroscopy of crystals. Also, the study of physical processes in EPR masers (population inversion, saturation effects, transient processes, etc.) stimulated research into relaxation processes in paramagnetic doped crystals.

These studies have provided fundamental insights into EPR spectroscopy and the physics of relaxation phenomena.

Much was learned about the fine and hyperfine structure of the EPR spectra of different crystal classes, more understanding was gained about EPR-line-broadening mechanisms, and new spin – lattice and spin – spin relaxation processes were discovered.

Of particular note are the results concerning relaxation processes in multi-level systems; as noted in Section 2, they are of fundamental importance to the physics of masers in that they determine the possibility of population inversion in a particular material while controlling saturation and transition processes in amplifiers and generators.

Specific aspects of the spin – lattice relaxation in multilevel systems — resonance relaxation processes via intermediate levels — were discovered and analyzed in Refs [21, 22]. Such processes are characteristic of multilevel systems and are observed at low temperatures in many crystals that have iron-group and rare-earth-group paramagnetic ions in their composition.

In this context, interpretations of processes such as the two-step resonance (Orbach et al. [21], 1961), a Raman-type process (Aminov [24], 1962), and a special case of the direct process (Manenkov and Prokhorov [23], 1962) were developed. The last interpretation, commonly accepted as correct, is schematically illustrated for a three-level spin system in Fig. 3, where wavy lines represent transitions between the corresponding energy levels due to spin – phonon interactions.

Kinetic analysis shows [23] that in the general case, level populations in such a system relax in a very complex (nonexponential) fashion depending on the probabilities of all the transitions involved.

In some special cases, however, population relaxation rates may relate among themselves in a single-exponential way. For example, if the relaxation transition probabilities between the two lower levels are much lower than those between the remaining level pairs (w_{12}, $w_{21} \ll w_{23}$, w_{32}, w_{13}, w_{31}), then the system uses level 3 to relax. In another case, when direct relaxation transitions between the upper levels 2 and 3 are much less probable than those between the other level pairs (w_{23}, $w_{32} \ll w_{12}$, w_{21}, w_{13}, w_{13}), level 3 is one through which populations of levels 2 and 3 relax.

Studies of spin – spin relaxation have revealed a variety of multispin cross-relaxation processes that are due to spin – spin interactions, including resonant, harmonic, and combinational [25].

Referring to the illustration in Fig. 4, if the energy differences between the level pairs a, b and a′, b′ satisfy the condition $\Delta E_{ab} \approx \Delta E_{a'b'}$, then spin – spin interactions in such a system give rise to resonant energy exchange between these level pairs (resonant cross relaxation).

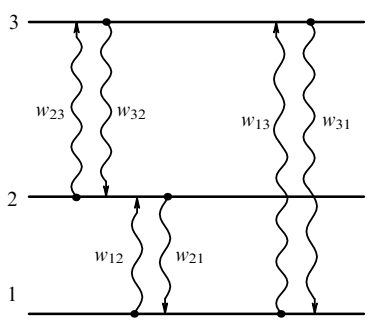

Figure 3. A three-level spin system. Wavy lines represent transitions due to spin – lattice interactions, with probabilities w_{ik} ($i, k = 1, 2, 3$).

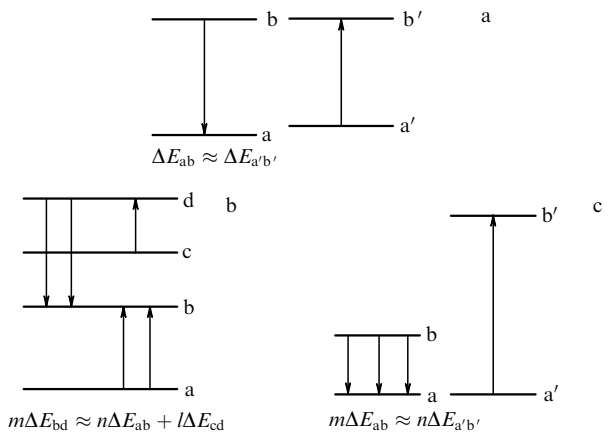

Figure 4. Cross relaxation in multilevel spin systems. Arrows indicate transitions due to spin–spin interactions for the resonant (a), combinational (b), and harmonic (c) cross relaxation. m, n, l are the numbers of spins participating in a corresponding cross relaxation process. Panel b, $m = 2$, $n = 2$, $l = 1$; panel c, $m = 3$, $n = 1$.

For other relations between the level energies, such as $m\,\Delta E_{ab} \approx n\,\Delta E_{a'b'}$ or $m\,\Delta E_{bd} \approx n\,\Delta E_{ab} + l\,\Delta E_{cd}$ (where m, n, l are the numbers of spins participating in the corresponding processes), the harmonic or combinational cross relaxation may occur, respectively.

Cross relaxation processes manifest themselves at high concentrations of paramagnetic ions (when spin–spin interactions are important) and appreciably enhance or reduce the possibility and degree of population inversion — and hence the efficiency of quantum amplifiers and generators.

We note that cross relaxation processes are of as much importance to EPR masers as they are to optical quantum amplifiers and generators (lasers), which use doped ion crystals as active media. We note here that cross relaxation processes in optical media have been insufficiently studied. This is especially true for combinational processes. The author knows of no reported observations of such processes in the optical range, although they have been definitely observed in the microwave range (in the system of spin levels of Cr^{3+} ions in ruby [25]).

5. From EPR maser to lasers

The development of microwave EPR masers encouraged extending quantum electronics principles to shorter wavelengths. That the first optical maser (i.e., laser) was realized on a ruby crystal (Maiman [26], 1960) was no chance occurrence. By that time, the physics of the microwave ruby maser was a well-developed field, and optical absorption and luminescence spectra of ruby had been studied in detail [27]. We also note the similarity in structure between the ground and excited states of ruby's Cr^{3+} ions — the states that determine EPR microwave spectra and optical spectra, respectively.

Importantly, preceding the first laser was an open-type cavity (mirror disk-shaped design) for molecular amplifiers and generators (masers) at submillimeter wavelengths (Prokhorov [28], 1958) — a key achievement that also had a major stimulating effect on research into short-wavelength masers. The first experimental studies of the disk cavity (in the millimeter wavelength range) showed that the open-type cavity allows obtaining high quality factors.

The success of the first ruby laser spurred activity in the field, triggering the effort to create solid-state lasers using doped crystals and glasses and to develop new active laser materials (atomic and molecular gases, semiconductors, liquids, etc.).

EPR and EPR maser studies largely shaped the way in which research into the physics of solid-state lasers developed in such areas as the spectroscopy of impurity ions in crystals, growth of large, optically homogeneous laser crystals, and lasing regimes (e.g., transients). We note that ruby played an especially important role in those studies and is in fact the mainstay QE material, from which highly efficient lasers and microwave masers are made. It is in ruby masers and lasers that physical processes have been most extensively studied, serving as models for describing solid-state masers and lasers in other crystals.

6. Some historical context

As any field of science, EPR and quantum electronics have their own complex history of ideas being unwillingly accepted and slowly recognized. What follows is some of my insider stories about the discoverers of EPR and QE.

E K Zavoisky's story

Historically, the discovery of the EPR phenomenon by E K Zavoisky in 1944 was made in a laboratory at Kazan State University. His first observations of the resonant absorption of electromagnetic radiation, Zavoisky said, were met with skepticism by his immediate colleagues at the Physics Department, and he therefore decided to conduct a special experiment to convince them that this was indeed a resonance effect, and a big one at that. The amplifier's output signal detecting the absorption effect was sent via a power amplifier to a dynamic loudspeaker mounted in the corridor, and Zavoisky then varied the magnetic field of the electromagnet within which the absorption cell of the EPR spectrometer was placed. The trick was that the loudspeaker produced a large volume of sound when the magnetic field was tuned to the electron paramagnetic resonance.

Zavoisky also said that some scientists — including such greats as Landau — did not think too much of his experiment and failed to recognize it as a major discovery — the reason, perhaps, that, even though repeatedly recommended, the discovery never won the Nobel Prize.

***A M Prokhorov* (*story excerpt*)**

"Sometimes Basov and I got a 'you guys are nuts' comment when explaining the idea."

***E L Feinberg* (*talk excerpt*)**

"Nothing fundamentally new here: stimulated emission was predicted by Einstein." (At the FIAN Academic Council session, commenting on the talk by Prokhorov on the idea of using stimulated emission to generate electromagnetic radiation).

***C Townes* (*story excerpt*)**

The ideas of QE took some time to gain acceptance even by such great scientists as Niels Bohr and von Neumann. C Townes [30] remembers their immediate reaction to the idea of a quantum generator as told by him. "You must be misunderstanding something," said Bohr. Von Neumann

Figure 5. EPR pioneers: E K Zavoisky (center), S A Altshuler (left), and B M Kozyrev (1968).

Figure 6. Quantum electronics pioneers. Left to right: A M Prokhorov, C Townes, N G Basov (1965).

first reacted in much the same way, "Oh, no, that can't be right. You must be misunderstanding something," but came back in 15 minutes to say, "Hey, you are right."

These two episodes are followed in Townes's book by a passage from Arthur Clarke — the kind of commentary which, the reader may agree, needs no comment:

"People go through four stages before revolutionary development:

1) It's nonsense, don't waste my time.

2) It's interesting, but not important.

3) I always said it was a good idea.

4) I thought of it first."

Instead of a conclusion, I hope two historical photographs featuring the pioneers of paramagnetic resonance and quantum electronics will not be out of place (Figs 5, 6).

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Magnetic resonance modes in spin-gap magnets

A I Smirnov

In some dielectric crystals with the antiferromagnetic exchange interaction, the projections of spin moments fail to show magnetic ordering even at temperatures approaching absolute zero. Such crystals came to be known as collective paramagnets or spin liquids. Examples of magnetic structures whose spin-liquid ground state is stable to perturbations are quasi-one-dimensional antiferromagnets containing chains