

Zeldovich's spin physics

(on the 110th anniversary of the birth of Yakov Borisovich Zeldovich)

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Abstract. We discuss Zeldovich's work in, ideas on, and findings in the physics of atomic–molecular magnetism and spectroscopy, the physics of angular momentum of electrons and nuclei, spin physics, and the physics of isotopes.

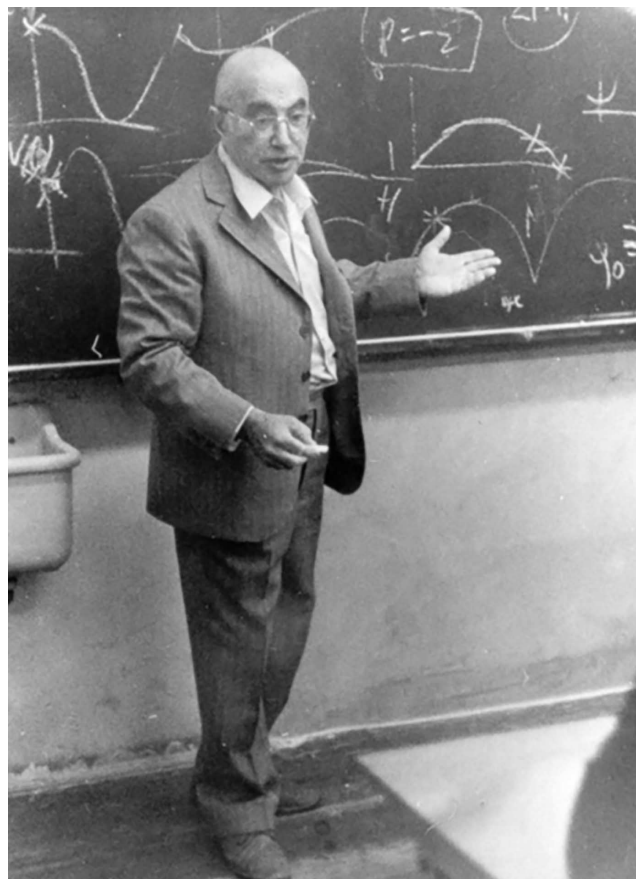
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From the Editorial Board

March 8, 2024 marked the 110th anniversary of the birth of Yakov Borisovich Zeldovich, an outstanding physicist and member of the Editorial Board of *Physics–Uspekhi* from 1964 until his death in 1987. In 2014, the year of Zeldovich's centenary, two special issues of *Physics–Uspekhi* were published that contained reviews and articles devoted to the development of his scientific heritage (see [2–15]). Another 10 years have now passed. Zeldovich's name continues to live in science in the development of the most modern areas of physics (see [16]). We decided to celebrate the 110th anniversary of Zeldovich's birth by publishing the following short but vivid note.

Zeldovich was multifaceted. His thinking was universal. It extended over many scientific fields and universally introduced new, often unexpected, unforeseen ideas, views, and opinions. He was a leader in the physics of combustion and explosion, in the theory of chain reactions (both chemical and nuclear), in nuclear physics and nuclear technology, in the physics of shock waves and detonation, in hydrodynamics, and in astrophysics and cosmology. This work by Zeldovich has not become obsolete and enjoys worldwide recognition. Less known are his ideas and findings in the physics of atomic–molecular magnetism and spectroscopy, in the physics of angular momentum of electrons and nuclei, that is, in spin physics, which is related to the physics of isotopes.

The fundamental importance of isotopes for our understanding of atomic and molecular phenomena and for



Yakov Borisovich Zeldovich
(08.03.1914–02.12.1987)

comprehending their mechanisms is well known. Three isotope effects are known to correspond to three fundamental properties of atomic nuclei: mass, volume, and spin (and the related magnetic moment). The most popular one, with a history spanning nearly a century, is the mass-dependent isotope effect; its magnitude is determined by the difference between the kinetic and vibrational energies of isotopic molecules. The magnitude of the nuclear-volume isotope effect is determined by the differences among the energies of the outer electrons of isotopic nuclei that differ in volume (due to deviations from the point symmetry of the potential, depending on the volume of the nucleus).

But the most unusual and diverse isotopic effects are induced by nuclear spin. Nuclear spin-selective optical excitation of atoms was proposed by Zeldovich and Sobelman in 1975 [17]. It goes back to the problem of laser separation of magnetic and nonmagnetic isotopes, i.e.,

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Yakov Borisovich Zeldovich and Anatoly Leonidovich Buchachenko at a conference on combustion (Tashkent, September 1987).

isotopes that have different nuclear spins. The simplest and most vivid example is the transition between electron levels in atoms with the total angular momentum $J = 0$; for the nuclear spin $I = 0$, this excitation is strictly forbidden, but, for a nonzero spin $I \neq 0$, the prohibition is lifted. Thus, in atoms with a closed ns^2 electron shell configuration (Ca, Zn, Mg, Hg, Cd, and others), the $ns^2 - nsnp$ ($^1S_0 - ^3P_0$) excitation is forbidden in spinless isotopes, but for atoms with spin-carrying nuclei, the probability of an electron transition (either absorption or emission) is not zero, because the hyperfine electron–nucleus interaction mixes the 1P_1 and 3P_0 states and makes the $^1S_0 - ^3P_0$ transition spin-allowed. Its probability is proportional to the coefficient $a[I(I+1)]^{1/2} \Delta E^{-1}$, where a is the hyperfine (Fermi) coupling constant in the 3P_0 state, I is the nuclear spin, and ΔE is the energy gap between the excited states 1P_1 and 3P_0 . Such transitions were observed experimentally in the spectroscopy of nuclear-spin atoms $^{199,201}\text{Hg}$ (2655.8 Å), but not in spinless atoms $^{200,202}\text{Hg}$. This effect may be interesting as a means of separating magnetic and nonmagnetic isotopes; perhaps it operates in space physics and chemistry.

Another effect of spin physics predicted by Zeldovich and Maksimov [18] relates to the diffusion of isotopic molecules in gases: they showed that, at low pressure, the diffusion coefficients depend on the nucleus spin. The idea lies in the conservation of the rotational angular momentum in molecules with spinless nuclei, with the result that the diffusion coefficient is determined by averaging the free flight paths over collision cross sections. In molecules with nuclear spin isotopes, rotational angular momentum is not conserved during flight due to the spin–rotation coupling $H_{IJ} = ICJ$, where I is the nuclear spin, J is the rotational angular momentum, and C is the spin–rotation coupling tensor. This effect is similar to the well-known Senftleben effect: the

dependence of the kinetic coefficients of molecular gases on the external magnetic field. The essence of the effect is related to the fact that nuclear spin violates the conservation of the angular momentum K of a molecule during its free flight between collisions. In the absence of nuclear spin, the diffusion coefficient is proportional to the mean free path averaged over different orientations of the angular momentum of the molecule. But in the presence of spin, the flying molecule tumbles and its angular momentum does not remain constant over the free flight path. In that case, the collision cross sections are to be averaged over free flight paths. Therefore, the scattering cross section is first averaged over the angular momentum directions, and then the mean free path is calculated given this averaged cross section. The results of these two physically different procedures are not identical: the diffusion coefficients of molecules with spinning and spinless nuclei are different.

Zeldovich proposed that this idea be used for the fractionation of nuclear isomers, that is, nuclei with the same masses and magnetic moments. Comparing the nuclear angular momentum precession frequencies (of the order of 10^7 Hz) with the frequencies of molecular collisions, he formulated the conditions for the detection and scale of this effect as a function of pressure.

Nuclear spin is of greatest significance in chemical physics, in the processes of electron–nucleus transformations that are classified as chemical, even though the underlying mechanisms pertain solely to the physics of the quantum electricity of atoms and molecules [19]; there, the Coulomb repulsion of electrons depends on their spin (singlet–triplet splitting) and they do not fall onto nuclei. The fundamental consequence of this physics is spin prohibition: all chemical reactions are selective with respect to electron spin, being allowed only for those spin states of reactants whose total spin is identical to the spin of the products; spin-changing processes are forbidden. The prohibition is universal, and it introduces magnetic (Zeeman and hyperfine) interactions into chemistry. While negligible compared with chemical energy, magnetic interactions are the only ones capable of changing the spin of reactants and switching reactions between spin-allowed and spin-forbidden channels to control the direction and rate of reactions.

The first remarkable consequence of the spin prohibition was the chemically induced polarization of nuclei, that is, the creation of super-equilibrium populations of nuclear-Zeeman levels. Molecules with oriented nuclei are selected by nuclear spin via the hyperfine interaction. Negative polarization is of particular interest: it results in excessively populating the upper Zeeman level. When the excess polarization exceeds the generation threshold, the chemically reacting system becomes a chemical maser — a chemically pumped quantum generator [20]. Zeldovich proposed regarding this phenomenon as a basis for chemical radiophysics. An example of radio frequency emission from a chemical maser is shown in Fig. 1; nuclear-polarized molecules are produced by photolysis of a quinone solution in the probe of an NMR spectrometer.

Another major consequence of spin prohibition is the magnetic isotope effect, the first isotope effect that distinguishes isotopes by nuclear spin and magnetic moment rather than by mass [21]. It was discovered in 1976 and was called the magnetic isotope effect, because it is driven by magnetic electron–nucleus hyperfine interactions in paramagnetic particles, intermediate products of

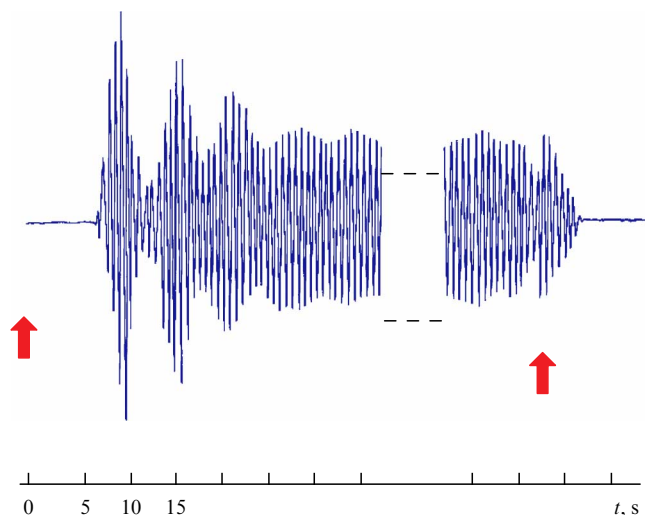


Figure 1. Chemically induced radiofrequency emission of protons emitted by quinone molecules. Signal is recorded at a frequency of 100 MHz. Arrows indicate the instants of turning photochemical pumping on and off.

1	H	He							
2	Li	Be	B	C	N	O	F	Ne	
3	Na	Mg	Al	Si	P	S	Cl	Ar	
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	
4	Cu	Zn	Ga	Ge	As	Se	Br	Kr	
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	
5	Ag	Cd	In	Sn	Sb	Te	I	Xe	
6	Cs	Ba	Lu	Hf	Ta	W	Re	Os	
6	Au	Hg	Tl	Pb	Bi	Po	At	Rn	
7	Fr	Ra	Lr	Rf	Db	Sg	Bh	Hs	
6		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd
7		Ac	Th	Pa	U	Np	Pu	Am	Cm

Figure 2. Part of Mendeleev's Periodic Table showing the elements for which the magnetic isotope effect has already been found (yellow).

reactions. The scale of the magnetic effect is one or two orders of magnitude greater than that of the classical mass-dependent isotope effect; it has already been discovered for the magnetic isotopes of carbon, oxygen, silicon, sulfur, germanium, tin, mercury, magnesium, calcium, zinc, and uranium (Fig. 2) in a variety of chemical and biochemical reactions, including those of biomedical and environmental significance [22, 23].

The magnetic isotope effect is the most striking phenomenon in spin physics. Demonstrating the dependence of reaction rates on the nuclear spin and nuclear magnetic moment of the reactants, it is a means of fractionating magnetic and nonmagnetic isotopes much more efficiently than by classical, technologically mastered methods. Zeldovich used to say that if the magnetic isotope effect had been

discovered earlier, the technologies for fractionating uranium isotopes would be different.

The magnetic isotope effect invokes new ideas on how to control chemical and biochemical reactions by selectively manipulating spin using magnetic fields and microwave pumping [22]. It even made a foray into medicine: the ability of magnetic isotopes of magnesium, calcium, and zinc to kill cancer cells and stimulate the enzymatic synthesis of ATP in living organisms was discovered [23].

The results of Zeldovich's work in spin physics are summarized in [24]; it is still widely cited today. He drew up the plan of the paper and wrote the first part, but did not see it in print format: it was published with his name inside a mourning border...

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