Double magnetoacoustic resonances in crystals

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The introduction contains an exposition of the basic principles of magnetic quantum acoutics and the possibility of its use to investigate the structure and the dynamics of solids. The distinguishing features of different double magnetoacoustic resonances (nuclear-nuclear, electron-nuclear, and electron-electron) are then considered. Results are presented of investigations of spin-phonon interactions, relaxation processes, and singularities of the structure in various classes of crystals, carried out with the aid of such resonances. This is followed by consideration of dynamic polarization of atomic nuclei with the aid of ultrasound. Particular attention is paid to the use of magnetoacoustic resonances for quantum amplification and generation of coherent phonons (acoustic masers) and for the development of quantum detectors for ultrasound.

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CONTENTS

1. INTRODUCTION

The study of the interactions between quantum systems in crystals and external alternating fields is one of the pressing problems of solid state physics. The most fundamental investigations carried out during the last two decades pertain to excitation of matter by electromagnetic waves of radio and optical frequency. Less investigated are processes connected with propagation of high-frequency (10^6–10^{11} Hz) elastic waves in solids and explainable only with quantum concepts. These include the interaction of ultrasonic waves with lattice vibrations or with conduction electrons in metals and semiconductors, and many other processes. In this paper we consider phenomena that pertain to resonant interaction of elastic waves with nuclear and electron spin systems in various classes of crystals.

The acoustic energy is transferred to the spin system by periodic perturbation of the internal interactions with the aid of the sound. Under resonance conditions, when the quantum energy of the elastic wave is equal to the difference between the energies of the spin levels, such a process gives rise to transitions of spins between different levels; an inverse process is also possible, in which the energy of the spin system can go over coherently into elastic waves. The absorption of acoustic energy by spin systems has the same character as spin-lattice relaxation, in which the energy of the spin system is transferred to the lattice via thermal phonons. The essential difference between these processes is that the relaxation, except at low temperatures, is via Raman scattering of phonons[1] in a wide frequency spectrum up to 10^{13} Hz, whereas quasimonochromatic phonons ($\Delta\omega \sim 10^2$ Hz) take part in the transfer of acoustic energy to the spins, the spectral energy density of the coherent acoustic phonons being 8–10 orders of magnitude higher than the density of the thermal phonons.

Acoustic magnetic resonance is the most direct method of studying spin-phonon interactions and obtaining information on relaxation processes in spin systems, and on the singularities of the structure and dynamics of intracrystalline electric and magnetic fields. Of particular value is the use of acoustic magnetic resonance in the study of metals, semiconductors, and superconductors, which are usually investigated with the aid of electromagnetic fields only in the surface layer of the material.

Acoustic magnetic resonance was predicted in 1952 by Al'tshuler.[2] In the same year, Kastler[3] considered the question of saturating spin systems with ultrasound. In the succeeding years, the theoretical study of various aspects of spin-phonon interactions was the subject of very many works, the main content of which is described in a number of reviews and monographs.[4–8]

The rate of the experimental investigations, however, was much slower, owing to the great complexity of the acoustic experiments; the main difficulty in acoustic nuclear magnetic resonance lies in observing nuclear absorption smaller by 4–6 orders of magnitude than the main "lattice" absorption; for acoustic electron paramagnetic resonance the difficulty lies in the generation and detection of elastic oscillations at fre-

quencies 10^9–10^{10} Hz.

At the present time, a direct method and indirect methods are used for the study of resonant absorption of acoustic energy by spin systems.

In the method of direct acoustic resonance, which is a mechanical analog of ordinary ESR and NMR, one measures directly the additional absorption of the acoustic power or the change of the phase velocity as the magnetic fields are swept smoothly through resonance. The magnitude of the spin-phonon interaction can be determined from the change in the absorption of the ultrasound at the instant of resonance.

However, owing to its insufficient sensitivity, the method of direct acoustic NMR did not find extensive use, and acoustic resonance with nuclear spins was observed by this method in a relatively small number of crystals, namely semiconductors of the III-V group,[9] alkali-halide crystals,[6] and certain metals.[10,11]

Successful observation of acoustic ESR in 1959[12] is due to primarily to the work of Baranskiĭ[13] on the development of methods for generating hypersonic oscillations in solids. Acoustic ESR was observed mainly on ions of the iron-group elements and rare-earth elements in diamagnetic matrices. A review of the results on acoustic ESR is contained in[1].

The method of direct acoustic ESR has been used to obtain data on spin-phonon coupling, and in some cases it is practically the only spectroscopic method for investigating impurity paramagnetic ions, for example, ions with an even number of electrons in crystals with symmetry lower than cubic.

The most informative and sensitive is the method of double magnetoacoustic resonances. The possibility of its use in acoustic magnetic resonance was first indicated by Kastler.[3,14]

The double-resonance method consists of simultaneously exciting a system of nuclear or electron spins by two alternating fields, one of which is acoustic and the other is of the electromagnetic type. To observe acoustically-induced spin transitions with the aid of double resonances it is necessary that the rate of the induced transition be larger than or comparable with the rates of all other processes that determine the stable distribution of the populations of the spin levels.

The results of recent investigations show that double resonances find extensive use in acoustic magnetic spectroscopy of atoms having a low concentration or having a weak spin-phonon coupling. The acoustic magnetic resonance is detected in this case by another spin system, contained in the same substance, but with a "stronger" NMR or ESR signal. Magnetoacoustic resonances make possible direct investigations of the mechanisms of spin-phonon interactions and all estimate the magnitudes of these interactions. Finally, the process of transferring acoustic energy from one spin system to another encompasses various mechanisms of internal interactions, and this makes possible a thorough study of the dynamics of spin systems. Just as the development of radio spectroscopy served as the impetus for the advent of quantum electronics, the investigation of spin-phonon interactions was the basis for the development of quantum amplifiers and generators for ultrasound–phonon masers.

In the succeeding sections we consider various types of double magnetoacoustic resonances and present the procedures and the results of the investigations by these methods.

2. METHODS OF OBSERVING DOUBLE MAGNETOACOUSTIC RESONANCES

From the point of view of the method, double magnetoacoustic experiments are similar in many respects to double resonances in radio spectroscopy. Widely current schemes are used for the registration of NMR and ESR signals.[15] The main difference lies in the use of special devices for the generation and the detection of the acoustic oscillations in the various frequency bands (10^6–10^{10} Hz). Coherent elastic oscillations are excited mainly with a piezo-transducer secured to one of the end faces of the investigated sample. The free transducer surface is located at the maximum of the electric field. At frequencies 10^6–10^8 Hz, plate-like transducers of quartz or lithium niobate are used. At higher frequencies, film transducers (CdS, ZnO) have come into use recently. The maximum intensity of the alternating electric field on the transducer is produced with the aid of metallic electrodes, and by cavity resonators at higher frequencies (10^9–10^{10} Hz).

When it comes to obtaining quantitative estimates of the spin-phonon interaction, a certain difficulty is raised by the measurement of the amplitude of the acoustic deformation. There are several methods for its determination.[6,16–19]

1) The method of measuring the equivalent electric impedance of the sample and of the transducer; 2) measurement of the lifetime of the coherent phonons; 3) an optical method using the diffraction of light waves by ultrasonic oscillations; 4) measurement of the induction emf produced by periodic displacement of a metallized end face of the sample in a constant magnetic field; 5) measurement of the displacement of the metallized end face of the sample using the dynamic capacitor made up of the end face of the sample and an immobile metallic plate. With the aid of these methods it is possible to measure a relative deformation amplitude up to 10^{-8} with accuracy 10–20% at frequencies 10^6–10^8 Hz. At higher frequencies, however, the measurement accuracy decreases abruptly.

Three independent systems are used to observe nuclear-nuclear acoustic resonance: excitation of ultrasound, measurement of the amplitude of the acoustic deformation, and monitoring the intensity of the NMR signal. One of the most widely used schemes of this type is shown in Fig. 1. The acoustic-saturation block makes possible both excitation of elastic waves at a fixed frequency and frequency scanning in the nuclear-transition bands at various rates.

One of the simplest types of double resonance is

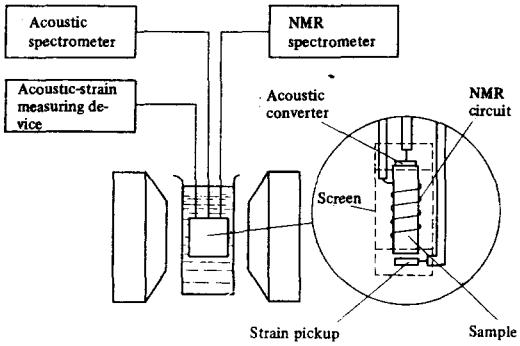

FIG. 1. Nuclear-nuclear acoustic resonance spectrometer.

acoustic saturation of NMR or ESR signals. Resonant acoustic oscillations of constant frequency cause transitions between spin levels; the resultant changes in the level populations are monitored against the change of the intensity of the NMR (ESR) signals by suitable continuous- or pulsed-radiation spectrometers. The acoustic-resonance line shape is studied by acoustically scanning the NMR (ESR) line at constant external magnetic field and at constant spectrometer frequency.

The general expression for acoustic saturation is

$$A/A_0 = (1 + w_a T_1)^{-1/r}, \qquad (1)$$

where A_0 is the initial signal intensity, A is the acoustically-induced intensity, $w_a \sim B G_{ij}^2 \varepsilon_j^2 g(\omega)$ is the probability of the acoustic transition, G_{ij} are the elements of the spin-phonon interaction tensor, ε_j are the elements of the acoustic strain tensor, B is a coefficient that depends on the magnitude of the spin and the magnetic quantum number m, $g(\omega)$ is the line-shape coefficient, T_1 is the spin-lattice relaxation time, and r ($\approx 1-3$) is determined by the structure of the acoustic waves in the sample. Thus, the spin-phonon coupling constant is determined from the degree of saturation (A/A_0) of the signal when the parameters in (1) are known.

The independent elements of the spin-phonon interaction tensor, the number which is determined by the symmetry of the crystal, can be determined for various directions of propagation of the elastic longitudinal and transverse waves relative to the direction of the external magnetic field and the crystallographic axes.

The acoustic saturation method is of considerable interest for the investigation of atoms having a nonequidistant spin spectrum, inasmuch as the process that brings such a system close to equilibrium is not described by a single relaxation parameter.

The difference $n_m = N_m - N_n$, between the populations of each pair of levels for acoustic saturation of a definite spin transition with $\Delta m = \pm 1$ or $\Delta m = \pm 2$, is described by a system of kinetic equations

$$\frac{d(n_m - n_0)}{dt} = \sum_i A_{mi}(n_i - n_0) + \sum_i B_{mi} n_i, \qquad (2)$$

where n_0 is the mean value of n_m in the absence of external perturbing fields. The coefficients A_{mi} and B_{mi} describe respectively the processes of the spin-lattice relaxation and of the external acoustic perturbation.

The use of analytic solutions of Eqs. (2) for spin values $J = 3/2$[20] and $J = 5/2$[21] makes it possible to estimate, from the degree of acoustic saturation of the various transitions, the probabilities of the acoustic and relaxation quadrupole and dipole transitions.

To increase the sensitivity of the acoustic ESR, a method of low-frequency nonresonant acoustic modulation of the ESR spectra was proposed.[22,23] Ultrasonic deformations via a spin-phonon interaction cause changes in the ESR spectrum, and this is analogous, to a certain extent, to the method of low-frequency modulation with an alternating magnetic field. But this procedure, while simple, is limited in its potential.

In substances with two sorts of spins, the procedure of observing acoustic NMR (ESR) of one sort of spins with the aid of another has much in common with double resonances in radio spectroscopy.[24] Initially the external magnetic field H_0 is set at a fixed value corresponding to the maximum of the absorption signal of the main atoms. The sweep of the acoustic saturation is then turned on in the region of the resonant transitions of the investigated spins and the change of the NMR (ESR) signal from the main atoms is recorded as a function of the ultrasound frequency.

The magnitude of the spin-phonon interactions can be estimated from the change of the NMR (ESR) signals from the main atoms following acoustic and electromagnetic saturation of the investigated atoms. The probabilities of magnetic ($\sim B_m \gamma H_1$) and acoustic ($\sim B_a G_{ij} \varepsilon_j$) transitions are compared at equal values of the saturation ($B_a G_{ij} \varepsilon_j \approx B_m \gamma H_1$; here γ is the gyromagnetic ratio and H_1 is the intensity of the electromagnetic field). For example, for the Al^{27}–Cr^{3+} system of nuclear spins in Al_2O_3[25] the constant G has a value 10^{-18} erg, and the equivalent ultrasound resonant absorption coefficient calculated from the value of G is $\alpha_n \sim 5 \times 10^{-11}$ cm^{-1}. This value of α_n is smaller by two orders of magnitude than the minimal ultrasound absorption coefficient previously determined by the method of direct acoustic resonance. To observe electron-nuclear resonance, one of the ends of the investigated sample is placed in the cavity resonator of the ESR spectrometer, and the ultrasound is excited by the method described above from the other end of the sample.

In magnetoacoustic electron-electron resonance, two independent microwave systems are used. The first is used to generate hypersonic waves that excite the transitions between the spin levels, and the other is used to monitor the change in the state of the electron spin system by the acoustic action. For a simultaneous excitation of the electron spin system by hypersonic and electromagnetic fields, one end of the sample is placed in a toroidal "acoustic" resonator and the other in the microwave resonator of the ESR spectrometer (Fig. 2).

It appears that the highest sensitivity can be obtained with the aid of optical detection of acoustic ESR, owing to the large values of the optical-transition quanta. The technique used to observe acoustic resonance is the same as for the optical detection of ESR spectra.[26]

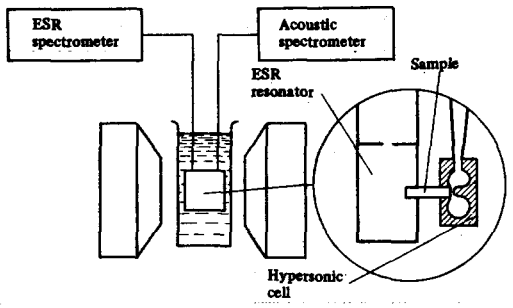

FIG. 2. Double acoustic electron-electron resonance spectrometer.

3. DOUBLE ACOUSTIC NUCLEAR-NUCLEAR RESONANCE

For dielectric crystals, the basic information on the mechanisms and constants of the spin-phonon coupling was obtained by the acoustic saturation method. Comparison of the experimental and theoretical data has shown that for nuclear spins with $J>1/2$ the spin-phonon interactions in impurity-free crystals is produced mainly by quadrupole interactions. The acoustic oscillations cause periodic displacements of the ions. As a result, alternating gradients of the intracrystalline electric field are produced and interact with the quadrupole moments of the nuclei. For nuclei with spin $J=1/2$ which have no quadrupole moment, the spin-phonon coupling is due to ultrasonic modulation of the magnetic dipole interaction between the nuclei. For piezoelectric crystals, a new "piezoelectromagnetic" mechanism was observed, based on the interaction of the magnetic moment of the nuclei with the "slow" electromagnetic wave accompanying the acoustic wave.[27]

It was noted earlier that spin-phonon interactions are closely related with relaxation mechanisms. It follows from (2) that various relaxation processes can be measured by the saturation method. An analysis of the solutions of (2) has shown that at $J>1$, for acoustic saturation of transitions with $\Delta m=\pm 1$ and ± 2, the difference between the populations of the spin levels can increase as well as decrease. As a rule, a decrease of the ratio n_m/n_0 takes place for transitions excited by sound, while an increase of n_m/n_0 is possible for transitions not excited by sound. If the spin-lattice relaxation is determined mainly by quadrupole interactions, then both an increase of the level population difference and population inversion are possible.[21]

An experimental verification of the theoretical premises has confirmed that acoustic saturation influences differently the individual population differences between the spin levels. In particular, enhancement of the intensity of the NMR signal as well as radiation induced by acoustic excitation of transitions with $\Delta m=\pm 2$, was observed for the nuclei Al^{27} in Al_2O_3[28] and Na^{23} in $NaNO_3$[29] and $NaClO_3$,[30] corresponding to a quadrupole type relaxation in these compounds.

A principally new approach to the problem of acoustic saturation was developed by Hamabusa and Yamaguchi.[31] They considered acoustic saturation of a nuclear spin system placed in a constant field H_0 and a sufficiently strong alternating magnetic field $2H_1\cos\omega t$, the frequency of which was close to a spin Larmor precession frequency $\omega_0=\gamma H_0$. In a coordinate system rotating with frequency ω around H_0, the spins are acted upon by a constant effective field H_e of intensity $\sqrt{(\omega_0-\omega)^2+\omega_1^2}/|\gamma|$, where $\omega_1=\gamma H_1$. Since $H_1\ll H_0$, the influence of the field H_1 on the orientation of the magnetic moments comes into play only at $\omega\sim\omega_0$, i.e., near resonance. Thus, the total change of the magnetization of the system near resonance will be determined by the intensity of the saturating field H_1. The field H_1 of frequency ω can be replaced by a field that is equivalent from the point of view of saturation of the spin system and has a frequency ω_1 equal to γH_1 for transitions with $\Delta m=\pm 1$ or to $2\gamma H_1$ ($\Delta m=\pm 2$). In the rotating coordinate system, the spin-lattice relaxation time T_1^* determined by measuring the decrease of the magnetization along the effective field will depend on the intensity of the saturation at the frequency ω_1

$$\frac{1}{T_1^*}=\frac{1}{T_1}+\frac{1}{T_a}, \tag{3}$$

where T_1 is the spin-lattice relaxation time in the absence of a strong alternating magnetic field, $1/T_a\sim G\varepsilon$. Experiments on acoustic saturation in a rotating coordinate system were carried out on Na^{23} nuclei in NaCl. Pulses of an alternating magnetic field of intensity H_1 were applied at the nuclear-spin Larmor precession frequency ($\omega/2\pi\sim 8$ MHz) in a field H_0. Acoustic saturation was produced at the frequency $\omega_1/2\pi\sim 4$ kHz. The acoustic-resonance line was swept by smooth variation of H_0 at constant values of ω and ω_1. The spin-phonon coupling constants were determined from the change of T_1^* near resonance (Fig. 3).

Even this very first experiment has shown that when acoustic saturation is used in a rotating coordinate system it is possible to decrease considerably the pump frequency (by 3–4 orders) and to increase appreciably the accuracy with which the spin-phonon coupling constant is measured.

One of the brilliant applications of the acoustic-saturation method was the corroboration of the concept of "spin temperature."[32] The Boltzmann distribution of the spin-level population was disturbed by saturating nuclear spins with equidistant spectrum (Na^{23} and Cl^{35} in NaCl) with the aid of ultrasound having double the Larmor frequency. If there is no spin-spin exchange

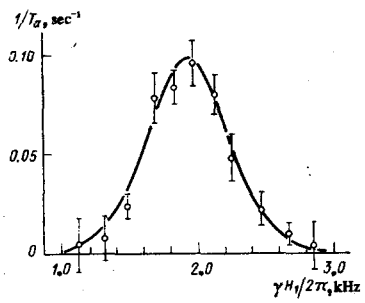

FIG. 3. Frequency dependence of the rate of acoustically induced nuclear relaxation.[31]

in the system, then the spin system preserves a definite value of the magnetization. However, owing to spin-spin interactions, a Boltzmann distribution of the populations is continuously maintained in the system and the magnetization of the acoustically-excited system decreases to zero. This is precisely what was observed in Proctor's experiments, with the establishment of the equilibrium described by a single characteristic time.

Further development of the "spin temperature" concept made it possible to describe thermodynamically a number of new phenomena that appear when a spin system is excited by an electromagnetic field. The most fruitful result of this approach was the segregation of the spin-spin interactions into a separate energy pool having its own heat capacity and temperature, different from the temperatures of the lattice and of the Zeeman systems.[33] The need for using the concept "spin-spin pool" in magnetoacoustic resonances is obvious, for in this case it is possible to provide an adequate thermodynamic explanation of the acoustic action that alters strongly the populations of the spin levels. The first to be investigated was the role of the electron dipole-dipole interactions following acoustic saturation of crystals with paramagnetic ions,[34] and then the concept of "dipole-dipole pool" was extended also to include pure nuclear spin systems.[35]

We consider now the results of a study of the dynamics of interactions of spin systems with one another and of a spin system with the lattice of a crystal in substances with two sorts of nuclear spins (pure dielectrics and dielectrics with paramagnetic impurities).

In the former case, the most complete data were obtained by Shutilov and co-workers in an impurity-free LiF crystal.[35] The ultrasonic excitations were excited in the crystal at the sum and difference precession frequencies of the nuclear spins of Li^7 and F^{19} in an external magnetic field. This gave rise to simultaneous joint transitions of both spins due to acoustic modulation of the magnetic dipole interaction between the Li^7 and F^{19} nuclei (acoustic-solid effect). According to the selection rules, the excited transitions are acoustically "allowed." The ultrasound at the difference frequency acts to increase the equilibrium static difference of the level populations for one type of spins (or to affect the dynamic polarization of the atomic nuclei), and action at the summary frequency leads to a decrease of the difference of the populations and

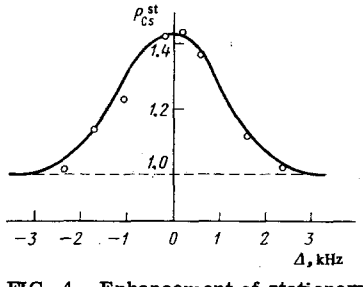

FIG. 4. Enhancement of stationary polarization of Cs^{133} nuclei in CsI by ultrasound.[36]

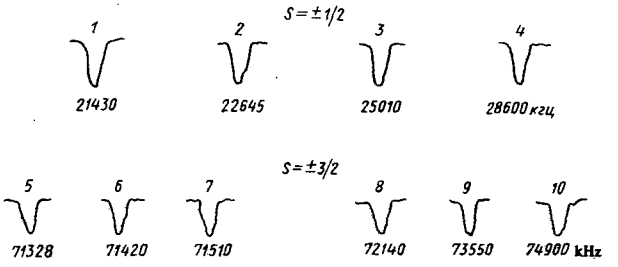

FIG. 5. Signals of double nuclear-nuclear magnetoacoustic resonance in ruby ($Cr^{53} \leftrightarrow Al^{27}$) ($\Delta m = \pm 1$).

further, to inversion of the spin-level populations. It has been established that the change of the polarization is connected with the behavior of the "unified pool" of the nuclear spin-spin interactions.

However, the possibilities of the acoustic-solid effects are not limited to the mechanism of modulation of the magnetic dipole-dipole interaction. In crystals with two sorts of nuclei I and S, in which one of the nuclear spins, S, is strongly coupled to the lattice, say by quadrupole interaction, dynamic polarization of the nuclei can be obtained by exciting "forbidden" acoustic transitions. The equilibrium population of a CsI single crystal was altered by saturation of forbidden transitions,[36] wherein the coupling of the lattice to the I^{27} nuclei was as strong ($T_1 \sim 0.01$ sec) and to the Cs^{133} nucleus anomalously weak ($T_1 \sim 500$ sec). The polarization was increased 1.5 times at the excitation frequency $\omega_2^- = (2\omega_I - \omega_{Cs})$ and correspondingly decreased at the frequency $\omega_2^+ = (2\omega_I + \omega_{Cs})$ (Fig. 4).

An investigation of the spin-phonon interaction curves for crystals with paramagnetic impurities was carried out in ruby (Al_2O_3: 0.07 at.% Cr^{3+}).[25]

The spin system of the host nuclei Al^{27} was used to detect the acoustic resonance between the hyperfine sublevels of the paramagnetic ion Cr^{3+}, due to the interaction with the nuclear moment of Cr^{53}. The acoustic-resonance lines for transitions with $\Delta m = \pm 1$ and ± 2 and electronic spin states $|\pm 1/2\rangle$ and $|\pm 3/2\rangle$ are shown in Fig. 5. The positions of the acoustic-resonance lines with $\Delta m = \pm 1$ coincides with the spectrum of the Cr^{53}–Al^{27} RF double resonance.[37] It should be noted that, in contrast to magnetic resonance, in acoustic resonance transitions are allowed not only between neighboring sublevels with $\Delta m = \pm 1$, but also with change $\Delta m = \pm 2$ of the magnetic quantum number. The change of the Al^{27} NMR signal intensity when the ultrasound passes through the center of the acoustic NMR resonance line of Cr^{53} nuclei amounted to 70–90% at a deformation amplitude 10^{-6}.

When the acoustic pumping frequency was shifted 10 kHz below the line center, an enhancement of the NMR signal of the Al^{27} nucleus by approximately a factor of two was observed, thus indicating an increase of the difference between the populations of the Al^{27} levels, i.e., the onset of dynamic polarization of nuclei.[38] At an ultrasound frequency 10 kHz higher than the resonant transition frequency, negative absorption was ob-

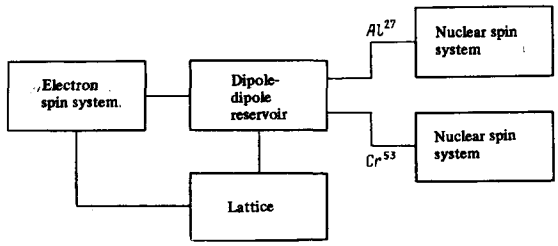

FIG. 6. Energy couplings between the nuclear and electron spin systems in ruby.

served, characterizing the inversion of the populations of the nuclear spin levels of Al^{27}.

In addition to the "forbidden" transitions with $\Delta m = \pm 2$, two-quantum transitions due to the summary action of two phonons of frequency ω_1 were observed in the Al^{27} system. Two-quantum acoustic excitation of the hyperfine transition $(1/2 \leftrightarrow 3/2)$ with frequency $\omega = 2\omega_1$ was revealed by the change of the intensity of the NMR signal of the Al^{27} nuclei. However, the probability of such a transition is much lower than that of the single-quantum transition.[39]

The nuclear subsystems of aluminum and chromium in ruby are not strongly coupled directly, since their resonance frequencies differ by almost an order of magnitude. However, an interaction between them can be realized via the dipole-dipole subsystem of the Cr^{3+} ions.[40] The over-all coupling scheme is shown in Fig. 6. The NMR frequencies of the Al^{27} and Cr^{53} nuclei fall in the interval of the electron spin-system relaxation rates. Calculations show[34] that acoustic saturation, owing to the coupling between the subsystems, equalizes the temperatures of the nuclear spins of Al^{27} and Cr^{53}. This changes the intensity of the NMR signal of the Al^{27} nuclei upon acoustic excitation of the spin transitions of the Cr^{53} nuclei. These experiments are the first confirmation of the role of the spin-spin pool in the dynamics of acoustic excitation of spin systems.

4. DOUBLE ACOUSTIC ELECTRON-NUCLEAR RESONANCE

The acoustic analog of double electron-nuclear resonance was theoretically considered by Deĭgen and Zheru.[41] They proposed to excite transitions between the hyperfine levels of a local center by acoustic oscillations, and to detect these transitions by means of their influence on the magnetic dipole transitions of the electron spins. Numerical estimates for F centers in KCl have shown that relatively low acoustic power (~ 2 W/cm^2) is needed to excite the nuclear spin transitions.

Simultaneous excitation of nuclear and electron spins by acoustic oscillations makes it possible, besides increasing the sensitivity and studying the mechanisms of the spin-phonon coupling, to investigate more fully the relaxation processes and the interactions between excited spin systems.

We shall consider the feasibility, in principle, of investigating such interactions in two different classes of crystals—dielectrics containing paramagnetic ions, and magnetically-ordered substances. These classes differ both in the mechanisms whereby the electronic and nuclear spin systems are coupled, and in their spin-phonon interactions.

In the former case, the investigated object was single-crystal Al_2O_3, containing 0.05 at.% of Cr^{3+} ions.[42,43] The sample was placed in a constant magnetic field and in two alternating fields: electric at the frequencies of the nuclear transitions of Al^{27}, and magnetic at the frequencies of the electronic spin transitions of the Cr^{3+} ions. To observe the acoustic NMR of the Al^{27}, the electron spin-system of Cr^{3+} was used. A change was observed in the intensity of the ESR signal following acoustic saturation at frequencies corresponding to nuclear-spin transitions with $\Delta m = \pm 1$ and $\Delta m = \pm 2$. The relative change in the ESR signal intensity at the centers of the double-resonance lines was ~10–15% at a relative deformation 3×10^{-7}. The lines of these resonances were approximately Gaussian with width $\sim 2 \times 10^4$ Hz at half-intensity. Figure 7 shows the double-acoustic electron-nuclear resonance lines and the values of the frequencies used to obtain the signals.

The experiments have shown that the interaction between the coherent acoustic phonons generated in the crystal lattice and the nuclear spin system proceeds via both quadrupole and dipole interactions. In the latter case, the acoustic energy is transferred to the nuclear spin system of the aluminum by modulation of the Cr^{3+}–Cr^{3+} magnetic dipole-dipole or exchange interactions. The participation of the paramagnetic spin system and the transfer of the energy of the acoustic oscillations to the nuclei has a virtual character and does not cause any change in the populations of the electron spin system of the ions. A comparison of the experimental results with theoretical calculations of the relaxation mechanisms and dynamic polarization of nuclei confirms the presence of a strong coupling between the dipole-dipole system the Cr^{3+} ions and the nuclear spin system of Al^{27}.[44,45] The following takes place during the course of the double magnetoacoustic resonance: microwave saturation of the ESR line raises the temperature of the Cr^{3+}–Cr^{3+} dipole-dipole pool relative to the unsaturated conditions, which in turn, via the Cr^{3+}–Cr^{3+} channel, lowers the temperature of the nuclear spin system. Acoustic excitation of the nuclear transitions decreases the spin-level population

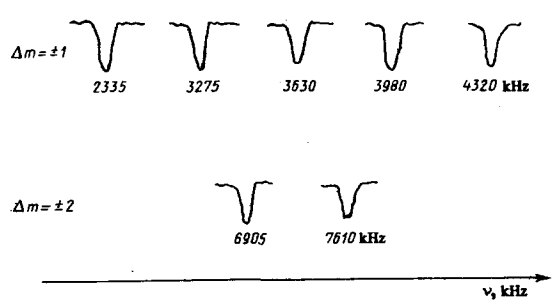

FIG. 7. Signals of double electron-nuclear magnetoacoustic resonance $A^{37} \leftrightarrow Cr^{3+}$ in ruby.

difference and thus leads to heating of the dipole-dipole system. In final analysis, the presence of a coupling between the electron Zeeman system of the Cr^{3+} ions and the system of the dipole-dipole interactions of the same ions raises the temperature of the electron spin system, as revealed by the decrease in the intensity of the ESR signal. The procedure described above was used also to observe Cr^{53}–Cr^{3+} double acoustic electron-nuclear resonance in ruby. The frequencies of the resonance peaks for the coupled Cr^{53} nuclei, obtained for electron-nuclear Cr^{53}–Cr^{3+} resonance, agree well with the analogous frequencies of Cr^{53} transitions determined from Cr^{53}–Al^{27} nuclear-nuclear resonance.[34]

Notice should be taken also of an "inverted" magnetoacoustic electron-nuclear resonance investigated[46] in single-crystal $CaF_2 : U^{4+}$. Since the ESR signal from the U^{4+} ions has a low intensity, acoustic ESR with the U^{4+} ions, which has much higher intensity, was used to detect the F^{19}–U^{4+} double resonance. The hypersonic paramagnetic absorption at constant H_0 was measured as a function of the frequency of the saturating radio-frequency field.

An increase or a decrease was observed in the amplitude of the hypersonic echo signal passing many times through the sample whenever the frequency of the alternating field was equal to the frequencies of the F^{19} nuclear transitions. The greatest changes in the acoustic absorption were observed in that case when the hypersonic pulses partially saturated the electronic transitions. These facts also confirm the hypothesis that the electron dipole-dipole interactions participate in the energy exchange between the nuclear and electronic spin systems. Broadening of the double-acoustic resonance F^{19}–U^{4+} is due to distortion of the local symmetry of the intracrystalline field by the acoustic oscillations. The investigation of the acoustic nuclear resonance in magnetically ordered crystals reveals a number of peculiarities. These include the presence of large internal fields at the nuclei, which shift the resonance frequencies, as well as nonlinear interaction of the electron and nuclear subsystems with each other and with the crystal lattice.[47–51] Indirect interactions of the nuclear spins via spin waves in the electron system become quite appreciable in cubic antiferromagnetics, which have by now been fully investigated with the aid of acoustic resonance.[52,53] The weak anisotropic field and the strong hyperfine field H_N of the above-mentioned substances cause a shift of the nuclear and electronic modes into a close frequency interval. The resonance frequencies for the electronic ($\omega_{e\pm}$) and nuclear ($\omega_{n\pm}$) modes (the + and – subscripts designate the modes that depend on the field H_0 and are independent of it, respectively), are given by[54]

$$\omega_{e\pm}^2 = \omega_{e_{1,2}}^2 + \omega_T^2, \quad \omega_{n\pm}^2 = \omega_N^2\left(1 - \frac{\omega_T^2}{\omega_{e\pm}^2}\right), \tag{4}$$

where $\omega_{e_{1,2}}$ are the antiferromagnetic resonance frequencies without allowance for the interactions with the nuclear spins, and ω_N is the hyperfine frequency.

The term ω_T, which causes the coupling of the unperturbed electronic and nuclear modes, is inversely proportional to the nuclear spin-system temperature. Whereas as at high temperatures this term is small, at low temperatures ($T_1 \sim 1$–$10\,^\circ$K) ω_T becomes of the order of ω_e and a dynamic coupling is produced between the two subsystems. This causes appreciable low-temperature shifts of the electron and nuclear frequencies. This phenomenon is used to observe NMR with the aid of double nuclear-antiferromagnetic resonance.[55] The saturation of the nuclear spin system by an external field raises its temperature, and this leads to a decrease in the low-temperature shift of the electronic (+) mode. The change in the frequency of the ω_{e+} mode is usually revealed by the shift of the resonant magnetic field H_0 at a constant antiferromagnetic-resonance frequency.

A theoretical analysis of the resonant absorption of acoustic energy by nuclei in magnetically-ordered substances has shown that such a process can be realized via two mechanisms.[51] In the first, periodic oscillations of the lattice cause, via the magnetoelastic coupling, oscillations of the electron spins and by the same token modulate the hyperfine field at the nuclei (the magnetostriction mechanism), while the participation of the electron system does not change its energy. In the second case, the lattice vibrations modulate the hyperfine or exchange electron-nuclear interactions directly. The dynamic shift of the electron and the nuclear frequencies was used to study double magnetoacoustic resonances in $RbMnF_3$ and $KMnF_3$ single crystals.[56] Saturation of the nuclear spin system by ultrasound at 500–670 MHz and power $\sim 10^{-2}$–10^{-3} W has led to a change in the resonant field $\Delta H_{0\max} \sim 300$ G of the (+) mode and to a change in the intensity of the (–) mode of the antiferromagnetic resonance observed at the frequency 9.5×10^9 Hz. So broad a nuclear-spin-pumping frequency interval, which causes a shift of the antiferromagnetic-resonance frequency, confirms the hypothesis[55] that the electron and nuclear spin modes become "entangled" at low temperatures. The connection between the pump frequency and the effective nuclear spin temperature T_n also agrees with this hypothesis (Fig. 8).

Another nonlinear effect is the power threshold of the acoustic pumping for the field-induced shift of the antiferromagnetic resonance, which was previously observed in electromagnetic pumping. It appears that

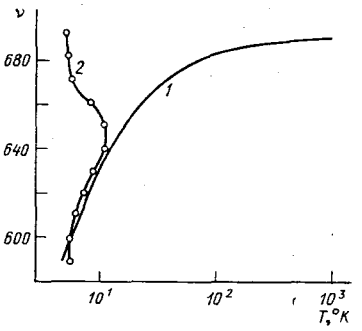

FIG. 8. Dependence of the acoustic-pump frequency ν (in MHz) on the nuclear spin temperature in $RBMnF_3$: 1—theoretical curve, 2—experimental curve at constant sound power.

acoustic saturation of the nuclear spins takes place first in those places where strong internal-filled gradients modulated by the acoustic oscillations exist.

It is known that NMR is observed in antiferromagnets only at one of the modes of the spectrum (ω_{n+}), inasmuch as for the other mode there is no amplification of the radio frequency field by modulation of the hyperfine fields.

In contrast to radio-frequency saturation, acoustic action makes it possible to investigate both nuclear-spin oscillation modes, since the gains of the acoustic oscillations are of the same order for both nuclear modes.

The equivalent coefficients (α_n) for the absorption of ultrasound by nuclear spins, determined by obtaining equal shifts of the field H_0 for antiferromagnetic resonance via acoustic and electromagnetic pumping, have values $\sim 10^2$ dB/cm. Calculations show that record values of α_n are produced by the combination of rather large NMR frequencies and the weak anisotropic field in cubic antiferromagnets. These values exceed by several orders of magnitude the values of α_n of dielectrics as well as of other magnetically-ordered substances.

5. ACOUSTIC DYNAMIC POLARIZATION OF ATOMIC NUCLEI

The phenomenon of dynamic polarization of atomic nuclei is used in nuclear physics to produce oriented nuclear targets and to obtain infralow temperatures. The attainment of stable dynamic polarization with the aid of acoustic saturation in solids, especially in large volumes of metals and semiconductors, could extend the region of applicability of this effect. Various methods were recently proposed for producing acoustic dynamic polarization. We have already mentioned[35,36] the increase of polarization by excitation of double nuclear-nuclear transitions with ultrasound. Another method of polarization was realized by adiabatic fast passage of the NMR by ultrasound.[57] Passage of the ultrasound frequency through spin resonance in a given magnetic field produced inverted population of the spin levels. Although these methods do not result in a high degree of polarization, they are quite promising for the investigation of internal interactions. A much higher polarization can be obtained by acoustic saturation of the electron spin system, using electron-nuclear interactions.

For dielectric crystals containing paramagnetic ions, acoustic pumping at frequencies $\omega_e \pm \omega_n$ produces a maximum polarization $p \sim \omega_e/\omega_n$, where ω_e and ω_n are respectively the electron and nuclear resonant frequencies. As shown in[58], for a system of nuclei with spin $J = 3/2$ in an axially-symmetrical crystal containing ions with an effective spin $S = 1/2$, the ratio of the polarizations by the dipole (D) and quadrupole (Q) coupling mechanisms is

$$p_D : p_{Q_1} : p_{Q_2} = 2\left[3 + 2\,\mathrm{sech}\left(\frac{\hbar\omega_e}{kT}\right)\right] : 10 : 5\left[1 + \mathrm{sech}\left(\frac{\hbar\omega_e}{kT}\right)\right]. \qquad (5)$$

For $\omega_e \sim 10$ GHz, $\omega_n \sim 10$ MHz, and $T = 0.5$ °K we have $p_D : p_{Q_1} : p_{Q_2} = 0.67 : 1 : 0.59$ at $p_{Q_1} = 633$.

An analysis of the mechanisms of ultrasound interaction with nuclei in metals[59] has shown that the transfer of the energy of the induced lattice vibrations to the spin system of the nuclei proceeds via a hyperfine interaction of the nuclei with the conduction electrons. In this case, the absorption of sound of frequency ω_a leads to polarization of the nuclei or to the acoustic analog of the Overhauser effect, provided that

$$\omega_a \gg \left(4kT + \frac{3}{2}\gamma H_0\right).$$

Acoustic dynamic polarization of Si^{29} nuclei in a strongly doped silicon single crystal was investigated by Hausser and Schweitzer.[60] Saturation of the electron spin levels of phosphorus ($T = 1.6$ °K) was obtained with 9×10^9 Hz hypersound. In this experiment, the hypersound was excited at one end face of the sample and the NMR-circuit coil was wound around the remainder of the sample in order to observe the change of the populations of the nuclear spin levels. Experiments on acoustic polarization in a sample containing 2×10^{19} phosphorus atoms have shown that the intensity of the NMR signal of Si^{29} is decreased by 1/3 following excitation by hypersound of ~ 10 μW power. In the authors' opinion, the main cause of the smallness of the effect is the insufficient power of the saturating acoustic field.

A higher degree of acoustic dynamic polarization of nuclei was obtained for Al^{27} nuclei in ruby.[61] Hypersound pulses of frequency 10^{10} Hz were introduced along the trigonal axis of the sample with the aid of a lithium niobate rod with metallized surfaces. This shielded the sample against the action of the powerful electromagnetic pulses exciting the piezoelectric transducer. The hypersound saturated the transitions between the spin levels ($-1/2 \leftrightarrow 3/2$) of the Cr^{3+} ions (concentration 0.01%). Acoustic saturation produced a tenfold change in the intensity of the NMR signal of Al^{27} at $T = 4.2$ °K. When H_0 was smoothly varied, amplification of the NMR signal was observed at first ($\gamma H_0 < \omega_e$). When the ESR line center was passed, a decrease of the intensity was observed, following by inversion of the populations of the spin levels of Al^{27} (Fig. 9). The distance between the intensity maxima of the NMR lines was 50 Oe. The time to restore the initial intensity after turning off the pump was 120 sec.

These results agree well with the theoretical concepts and confirm the feasibility of realizing acoustic polarization of nuclei.

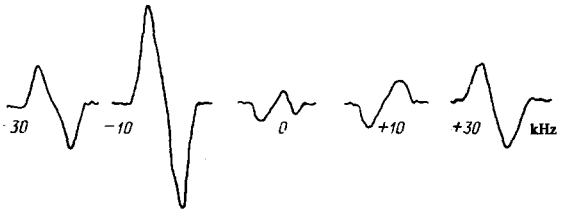

FIG. 9. Dynamic polarization of Al^{27} nuclei in ruby when the acoustic saturation is shifted in frequencies from the center of the ESR line of Cr^{3+}.

6. DOUBLE ACOUSTIC ELECTRON-ELECTRON RESONANCE

Double electron resonance was used in a number of studies[12,62-67] of the mechanisms of spin-phonon interactions in various crystals, as well as to investigate the singularities of the dynamics of spin systems under the influence of acoustic excitation. This method was used to observe acoustic resonance from Mn^{2+} ions and radiation centers in quartz,[12] Mn^{2+} and Fe^{3+} ions in corundum,[64] and Fe^{2+} and Fe^{3+} ions in MgO.[65] In the latter case, double resonance made it possible to distinguish between signals from Fe^{3+} and Fe^{2+} ions, even though the concentration of the latter was smaller by two orders of magnitude than that of the former.

The theoretical estimates and the experimental results show that in dielectric crystals containing paramagnetic ions of the intermediate groups, the interaction of the acoustic waves with the electron spin system is via the Van Vleck mechanism. In this case periodic lattice vibrations modulate the intracrystalline field at the locations of the paramagnetic ions. The orbital motion of the electron, which is sensitive to fluctuations of the crystal field, alter in turn, via the spin-orbit coupling, the states of the electron spins.

At high concentration of the paramagnetic ions, however, the Waller mechanism may become the effective one. Then the spin-phonon coupling is due to modulation of the spin-spin interactions by the lattice vibrations.

Worthy of attention, from the point of view of the study of the singularities of the spin-phonon interactions, are the studies of the excitation of the transition of one ion with simultaneous absorption of several phonons. The resonance condition for the n-quantum process is of the form $nh\nu = \gamma H_0$. Such resonant two-phonon transitions were observed initially for Fe^{2+} ions in MgO,[66] and multiphonon ($N = 2, 3, 4$) processes were observed for the transition $(1 \longleftrightarrow -1)$ of U^{4+} ions in CaF_2.[67] It follows from the angular dependence of the resonant frequencies and of the absorption intensity that acoustic resonant absorption is indeed due to n phonons of frequency ν, and not to one phonon of frequency $n\nu$. However, the nonlinear increase of the acoustic absorption following an appreciable increase of the hypersound power is apparently due to the onset of unharmonic oscillations with frequencies that are multiples of ν.

Acoustic excitation of various spin transitions in $Al_2O_3:Cr^{3+}$,[62] which increase or decrease selectively the intensities of the ESR lines, has made it possible to obtain additional information on the relaxation processes, particularly on the contribution made to them by phonons of different polarization.

The spin-lattice and spin-spin relaxation mechanisms can also be investigated by acoustic modulation of the ESR spectra in the interval of the rates of these relaxation processes ($w_a \sim T_1^{-1}$ and $w_a \sim T_2^{-2}$, where T_2 is the spin-spin relaxation time).[68]

Let us dwell in somewhat greater detail on optical detection of acoustic ESR in molecular crystals. The spin-phonon interactions in excited paramagnetic states is much weaker for organic molecules than for dielectrics containing paramagnetic ions, so that it is hardly possible to observe acoustic ESR by a direct method. However, the high sensitivity of optical detection has made it possible to observe the absorption of sound in the paramagnetic triplet state of tetrachlorobenzene in a zero magnetic field.[69] Optically detectable acoustic ESR was revealed by the change of the intensity of the phosphorescence at a wavelength 3782 Å with the hypersound frequency swept in the region of the $2E$ transition (1.7×10^9 Hz). A 30% change of the intensity corresponded to an acoustic power of ~ 100 μW in the sample. Investigations of acoustic ESR in molecular crystals has made it possible to study the anisotropy of the spin-lattice coupling and the contribution made to it by longitudinally- and transversely-polarized phonons.

7. ACOUSTIC MASERS AND QUANTUM DETECTORS FOR PHONONS

Progress in the research on double magnetoacoustic resonances has led logically to the study of the possibility of amplifying and generating monochromatic coherent phonons by spin systems and to the development of acoustic masers. The first theoretical papers by Kopvillem and Korepanov,[70] Townes,[71] and Kittel[72] dealt with a general quantum system, which was analogous to some degree to the system already used to amplify and generate photons.[73] Inversion of the spin-level population by an additional resonant field, say of electromagnetic nature, at sufficiently strong spin-phonon coupling should lead to an enhancement of both the thermal phonons and the externally generated monochromatic acoustic waves with frequencies corresponding to a definite spin transition. Phonon generation sets in upon self-excitation of the system because of the use of a high-Q acoustic resonator, the role of which, just as in solid-state lasers, is assumed by the sample itself.

Amplification of acoustic pulses was first obtained by Tucker in the electron spin system of the Cr^{3+} in ions. The experimental procedure is similar in many respects to the observation of double acoustic electron-electron resonances. To produce population inversion of the spin levels, one end of the cylindrical sample was placed in a microwave resonator. A quartz transducer was used to excite in the second end of the sample acoustic pulses at the frequency of the spin transition. At an electromagnetic pump power 40 mW and a frequency 9.3×10^9 Hz and $T = 4.2$ °K, the damping of the hypersonic waves was decreased 12% per centimeter of sample length in comparison in the case when the electromagnetic pump was turned off (Fig. 10).

This first experiment indicated a realistic possibility of using the principles of stimulated emission and absorption to excite spin systems by acoustic fields.

Phonon generation occurs when the inverse amplification greatly exceeds the total losses to damping of hypersound in the sample and in the coupling layer, and

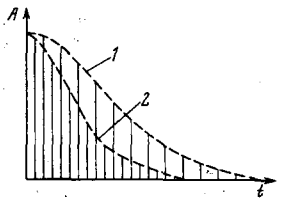

FIG. 10. Increase of the amplitude of acoustic echo pulses following electromagnetic pumping (1) in comparison with the usual damping (2).[74]

to dissipation in the surrounding medium. Indeed, Tucker[75] succeeded somewhat earlier, by using two-resonator installation, in generating phonons. Calculations have shown that to compensate for the losses in the system and to reach the generation threshold the required gain is 0.11 cm^{-1}. The total width of the generation band was 11 MHz, much less than the width of the transition line (~ 50 MHz). The acoustic-oscillation power in the ruby rod, determined by measuring the electromagnetic radiation from a quartz transducer, did not exceed 10^{-7} W.

A similar method was used in[76] to obtain quantum amplification on Ni^{2+} ions in Al_2O_3. The experiment was performed with a plane-parallel sample containing 5×10^{-4} % Ni^{2+} ions. The acoustic pulses were generated by a CdS film piezoelectric transducer at a frequency 9.4×10^9 Hz with electromagnetic saturation of a transition of frequency $\nu = 5.3\times10^{10}$ Hz. The spin-phonon interactions were several times stronger for Ni^{2+} ions than for Fe^{2+}, so that a gain of 0.013 dB/cm at $T = 2.2$ °K was observed. This gain exceeded the loss to hypersound damping in the sample, but no generation of acoustic oscillations could be obtained; in the authors' opinion this is due to the dissipation of the hypersound in the sample because the end faces were not parallel, the crystal structure was not perfect, and the ion concentration was low.

Shiren[77] obtained inverted populations by using the method of adiabatic fast passage of a magnetic field through resonant spin transitions with $\Delta m = \pm 1$ of Ni^{2+} and Fe^{2+} ions in MgO. After establishing the inversion of the levels $(1 \rightarrow -1)$, double photon-phonon transitions were excited by simultaneous action of electromagnetic and acoustic pulses ($\nu_{em} + \nu_q = \nu_{1\rightarrow-1}$). At $T = 1.6$ °K, an acoustic gain of 10 dB/cm was observed for Ni^{2+} and 24 dB/cm for Fe^{2+}. These values are somewhat lower than the theoretically calculated gain, owing to the incomplete inversion of the population and its destruction by the short spin-lattice relaxation times.

Considerable progress in the generation of coherent phonons was reached by Ganapolskiĭ and Makovetskiĭ[78,79] with the aid of an improved method of exciting acoustic oscillations and producing a low-loss hypersonic resonator (resonator $Q \sim 5\times10^5$) (Fig. 11). The populations of the spin levels of the Cr^{3+} ions in ruby was inverted by electromagnetic pumping at 2.3×10^{10} Hz. The result was a coherent spontaneous emission of phonons with intensity 10^{-6} W/cm^2 at a frequency 9.1×10^9 Hz ($T = 1.7$ °K). The frequency spectrum of the acoustic oscillations consists of individual narrow lines in each of which the radiation monocromaticity ($\Delta\nu/\nu$) is less than 2×10^{-6}. In the authors' opinion, the aggregate of the emission line corresponds to simultaneous generation at several neighboring acoustic modes of the hypersonic ruby resonator. It is suggested on the basis of the shape of the spectrum that the level width of the elementary spin excitation is $\Delta\nu \sim 3\times10^5$ Hz, which is smaller by two or three orders than the width of the inhomogeneous resonant ESR line of the Cr^{3+} ions in ruby. The lifetime of the coherent phonons, determined from the threshold gain ($\alpha_n \sim 0.55$ dB/cm) is 7×10^{-6} sec, which agrees with the analogous values obtained from the width of a ruby-resonator resonance curve and from the attenuation of the echo pulses in a resonator in the absence of a magnetic field.

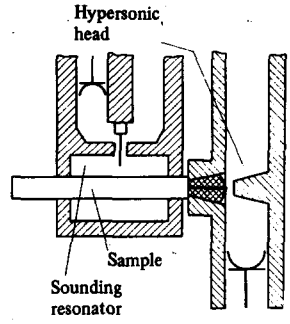

FIG. 11. Resonator for the excitation of hypersound.[79]

It follows from the described results that for an acoustic maser it is desirable to use samples with sufficiently small inherent acoustic damping, containing paramagnetic ions with large spin-phonon interactions. Substances that can be used in acoustic masers are listed in the table.[80] The spin-phonon coupling constants G_{ij} are given in cm^{-1}/strain unit.

It appears that the frequency limit of the action of acoustic masers can be greatly extended by excitation of spin transitions with laser pumping in substances containing optically active centers. A detailed calculation[81] carried out for laser excitation in $MgO:Cr^{3+}$ has shown that the generation of phonons with frequency up to 3×10^{12} Hz is perfectly realistic, and up to 8×10^{12} Hz in $CaF_2:Sm^{2+}$ crystals.

Soon after the first experiments on double magnetoacoustic resonances, suggestions were advanced to use them to detect acoustic phonons of low density.

A method of identifying acoustic oscillations in pulsed and continuous regimes with the aid of two-quantum transitions in a three-level system was proposed in[82]. A scheme with simultaneous absorption of a photon and a phonon makes it possible to use a variety of combinations of phonon frequencies ω_a and photon frequencies

Substance	Ion	G_{11}	G_{44}	$\sqrt{G_{16}^2 + [(G_{11}+G_{12})^2/4]}$
MgO	Ni^{2+}	57	50	—
MgO	Fe^{2+}	650	380	—
MgO	V^{3+}	1000	—	—
Al_2O_3	Fe^{2+}	90	—	—
Al_2O_3	Mn^{3+}	500	—	500
Al_2O_3	Cr^{4+}	—	—	60
Al_2O_3	Fe^{2+}	90	—	15
Al_2O_3	V^{3+}	55	—	140
$KMnF_3$	Ni^{2+}	58	39	—
$KMnF_3$	Fe^{2+}	1340	1000	—
CaF_2	U^{4+}	170	—	—

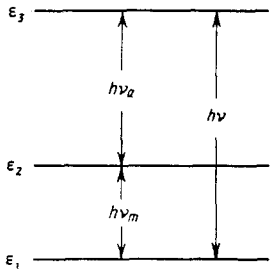

FIG. 12. Two-quantum transition scheme.

ω_m, provided that the condition $(\omega_m + \omega_a) = \omega$ is satisfied (Fig. 12), where ω is the frequency of the two-quantum transition. It is emphasized in[82] that two-quantum indication, with a sensitivity equal to that of the method of piezoelectric conversion, does not require phase coherence of the signal, and for Fe^{2+} ions in MgO hypersonic oscillations are observed in the 300 MHz band at a fixed electromagnetic field frequency.

An appreciable increase in sensitivity can be obtained in such a three-level scheme by using infralow temperatures, when practically all the spins are at the lower level (1).[83] The two upper levels (2 and 3) are populated by simultaneous acoustic and electromagnetic excitation of the transitions $(1 \leftrightarrow 2)$ and $(2 \leftrightarrow 3)$. The authors of[83] believe that it is possible to observe in a sample a flux of 1 phonon/sec at a temperature ~ 0.01 °K.

Acoustic saturation of the ESR (NMR) lines, which leads to a change in the spin-level populations, can be used, if the spin-phonon interaction constants are known, to measure the amplitude of the deformation or acoustic power. Use is made here of the proportionality of the saturation factor A/A_0 to the product $G_{ij}\varepsilon_j$. The minimum value of the observed deformation depends on the ratio $\Delta A/A_n$, where $\Delta A = A_0 - A$ and A_n is the overall noise of the measurement setup, referred to the input. For the previously investigated nuclei (for example In^{115} in InSb)[84] the value of ε_{min} is 10^{-6} at $T = 4.2$ °K in the frequency band 1–20 MHz.

To increase the sensitivity of the measurement by the acoustic saturation method, it was proposed to use dynamic polarization of nuclei, with the aid of which it is possible to increase the spin-level population difference by a factor ω_i/ω_n, and accordingly to increase the sensitivity of the quantum generator by the same factor. Measurement of the amplitudes of alternating deformations was performed on a ruby single crystal $(Al_2O_3 : 0.46\%\ Cr^{3+})$.[85] An electromagnetic field was used to saturate the $(-3/2 \leftrightarrow -1/2)$ transition of the Cr^{3+} ions at the frequency 3.6×10^{10} Hz. The change in the difference of the spin-level populations of the Al^{27} nuclei was determined by registering the change in the NMR signal. At an electromagnetic pump power ~ 5 mW and $T = 4.2$ °K, the change obtained in the population difference of the Al^{27} levels was larger by two orders than the equilibrium value. Acoustic oscillations at the frequency of the transitions of the nuclear spins of Al^{27} were then excited in the sample. The frequency range 3–20 MHz was covered by varying the magnitude and

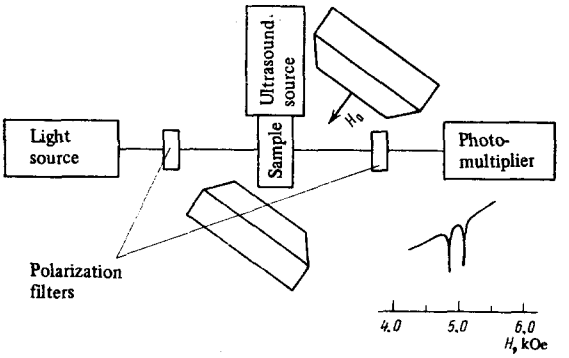

FIG. 13. Acoustic spin detector and acoustic-oscillation signals detected by optical dichroism in $CaF_2 : Tm^{2+}$.[86]

direction of the constant magnetic field H_0 relative to the crystal axis. At $\Delta A/A_n \sim 5$, the measured strain amplitudes were 7×10^{-9}, which is smaller by two orders of magnitude than the measured ε_{min} in the same sample, but in the absence of dynamic polarization of the nuclei. Since the degree of polarization is proportional to $h\nu/kT$ it is desirable to lower the temperature and to increase the pump frequency in order to increase the dynamic polarization. At $T \sim 10^{-1}$–10^{-2} °K and $\nu \sim 10^{11}$–10^{12} Hz, this method can be used to produce ultrasensitive acoustic strain gauges.

Acoustic waves in the microwave band were detected with the aid of a spin-phonon spectrometer by Anderson and Sabrinsky.[86] Tm^{2+} ions in CaF_2 served as the quantum detector. The change of the spin-level populations at frequencies 10 and 24 GHz were detected by the circular dichroism of the crystal (Fig. 13). Since the intensity of the circular polarization of a paramagnet depends on the spin population, what was measured in the experiment was the dependence of P on the number of spins at a given level N_i. For example, for a two-level system $P/P_0 = (N_1 - N_2)/(N_1 + N_2)$. At a temperature 1.4 °K, the minimum power of the monochromatic acoustic wave, as determined from the dichroism, is 10^{-11} W.

8. CONCLUSION

The data presented in this review demonstrate the extensive possibilities of a detailed investigation of the features of internal interactions in various classes of crystals by methods of double acoustomagnetic resonances. Further use and development of this method will undoubtedly extend the existing concepts concerning the structure and the dynamics of the intracrystalline fields, of relaxation processes, etc. One of the promising directions of this method is the use of phonon induction and echo,[87] as is evident by recently obtained first experimental results.[88] Of undoubted interest is the search for substances with record high spin-phonon interactions, which can be used as working media for acoustic masers, and also the development of new methods of generation and amplification of coherent acoustic oscillations. It appears that the effective sound generation by spin systems in the superradiance region can also find application here.[89,90]

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