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Influence of local anisotropy on the states and resonant properties of an optically oriented system of electron and nuclear spins of semiconductors

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A strong anisotropy has been observed in the optical orientation of the electron and nuclear spins in $Al_xGa_{1-x}As$ ($x \approx 0.25$) crystals at 77 K. Depending on the angle φ between the [110] crystallographic axis and the transverse magnetic field H_x (the light propagates in the z direction), different states of the spin system and transitions between them are observed. At small angles φ there is a range of fields in which two stable polarization states exist. The width and the position of the boundaries $H_{x1}{}^b$ and $H_{x2}{}^b$ of this range change abruptly (by several dozen oersteds) when φ changes by several degrees. The jumplike transitions from one stable state to another can be initiated by a change in the field H_x near H_{x1} or H_{x2}, by turning off the light for a short time, or by turning on an alternating field $H_{1y}^{\sim}$ at the nuclear magnetic resonance (NMR) frequency. With increasing φ, the width of the hysteresis region decreases, and at $\varphi \approx$ 3–5° undamped relaxation oscillations of the polarization set in, with a period from several to about 10 seconds. The period and the waveform of the oscillations depend on the angle and on the field H_x, and can vary under NMR conditions (at $\varphi = 45°$ (the [100] axis coincides with H_x) the behavior of the polarization is well described within the framework of the cooling of the nuclear spin system in the field of the electrons oriented by the light. Optical observation of NMR of As^{75} nuclei has made it possible to determine the role of the local anisotropy that arises in the semiconductor as a result of violation of the cubic symmetry when the Ga atoms of the initial GaAs crystal is replaced by Al. The angular dependences of the resonant frequencies include two branches that vary like $3\gamma H_x \cos\psi\cos\varphi$ and $3\gamma H_x \cos\psi\sin\varphi$, where ψ is the angle between the axes [111] and [110] and γ is the nuclear gyromagnetic ratios. These branches correspond to different positions of the substitutional Al atoms in the unit cell. Resonances have been observed at the subharmonics that correspond to multiquantum transitions. The probabilities of one-, two-, and three-quantum transitions ($\pm 3/2 \to \mp 3/2$) are compared. The role played by nuclear quadrupoles in the formation of the effective magnetic field that acts on the electron spins is discussed.

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1. INTRODUCTION

When circularly polarized light is absorbed in a semiconductor, the angular momentum of the light quanta can be transferred to the nuclear spin system. In an external magnetic field $H \gg H_L$, where H_L is the local field of the nuclei, it is possible to accumulate an appreciable nuclear polarization along that component of the field H which is parallel to the exciting-light beam.[1-4] This effect can be well described within the framework of the ordinary Overhauser effect. In weak magnetic fields, an appreciable lowering of the spin temperature of the nuclei by light is possible.[5-8] This leads to a number of characteristic singularities in the behavior of the optically oriented electrons. Thus, for example, in a weak magnetic field perpendicular to the light beam, the electron polarization can undergo strong changes.[7-9] This effect is due to the unique enhancement of the field by the hyperfine interaction. The electron spins are acted upon by the effective fields of the oriented nuclei, and this field can greatly exceed the external field. This situation is the converse of the standard situation in electron paramagnetic resonance (EPR) and electron-nuclear double resonance (ENDOR), where the hyperfine fields at the electrons are much weaker than the external field.

The anisotropy of the hyperfine interaction should lead to anisotropy of the spin orientation of the electrons. Until recently there were no investigations of the anisotropy effects in optical orientation of spin in semiconductor. Our earlier paper[10] seems to be the first communication on this subject. As will be shown below,

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the anisotropy of the crystal gives rise to a group of strong effects that manifest themselves in the optical channel when the circular polarization of luminescence is investigated.

The lowering of the cubic symmetry following substitution of part of the atoms of cubic crystals that contain nuclei with spin $I > 1/2$ creates conditions for manifestations of quadrupole interaction. Such a situation arises in the investigation of GaAlAs solid solutions wherein some of the gallium atoms of the original GaAs crystal are replaced by aluminum atoms. All the nuclei of the GaAlAs lattice are paramagnetic and have a spin $I = 3/2$ (with the exception of ^{27}Al whose spin is 5/2). The quadrupole splitting of the nuclear levels under conditions of optical orientation leads to the appearance of additional effective magnetic fields that act on the electron spins. The high sensitivity of the electron polarization to the hyperfine fields in the case of optical orientation in weak external field makes it possible to observe optically forbidden transitions in nuclear magnetic resonance (NMR). Allowance for the quadrupole interaction explains the behavior of a nuclear spin system in an external field $H > H_L$. In this region, the model wherein the spin system of the nuclei is cooled in the field of optically oriented electrons[7] underestimates the nuclear fields at the electrons. The hyperfine fields connected with the quadrupole splitting lead to a strong anisotropy of the behavior of the electron spin polarization when the orientation of the external field relative to the crystallographic axes is changed. The interaction between the electron and nuclear spin systems under optical orientation in a magnetic field perpendicular to the exciting light beam can lead to the appearance of regions where the average electron spin becomes a multiply valued function of the external field and of the crystal orientation. This manifests itself in the experiment in jumplike changes of the circular polarization of the luminescence at the boundaries of the regions of multiple values—a sort of optical analog of the "switching" effect. Slow undamped oscillations of the polarization are observed near these regions.

A complete theory describing the phenomenon to its full extent has not yet been developed. In this article we confine ourselves to an exposition of the principal experimental results and to their qualitative discussion.

2. EXPERIMENT AND PRINCIPAL RESULTS

The behavior of a system of oriented electron and nuclear spins can be easily traced by measuring the degree ρ of the circular polarization of the luminescence. The quantity ρ in the investigated crystals is numerically equal to the projection $\langle S_z \rangle$ of the average spin $\langle S \rangle$ of the electrons on the observation direction. The polarization of the nuclei is not measured directly in this case. It is possible, however, to realize conditions such that ρ turns out to be very sensitive to the state of the nuclear spin system. This situation arises when the electron spins are optically oriented in an external magnetic field H_x perpendicular to the exciting beam. When superimposed on the external field, the effective field $\mathbf{H}_N$ of the nuclei changes the precession rate of the electron spin as well as the experimentally observed value of ρ. The longer the lifetimes and the spin relaxation of the nonequilibrium electrons, the weaker the fields that cause a noticeable change of ρ and the more sensitive the circular polarization of the luminescence to the polarization of the nuclei.

We present below the results of an investigation of epitaxial samples of p-type $Al_xGa_{1-x}As$ crystals, for which the changes of the magnetic fields amounting to several or several dozen oersteds greatly influence the value of ρ. This corresponds to a sensitivity on the order of 10^{-4} to the degree of nuclear polarization.

1. Experimental setup

A simplified diagram of the experiment is shown in Fig. 1. The circularly polarized He-Ne laser beam produces along the z axis an optical orientation that coincides with the normal to the (100) plane of the crystal. By rotating the crystal around the z axis we can obtain the functions (H_x) for different angles φ between the [110] axis and the external field H_x directed along the x axis. An alternating magnetic field $H_{1y}^{\sim}$ applied along the y axis was used to excite low-frequency resonant transitions in the spin system of the nuclei.

The resonances were determined from the change of ρ following the action on the effective field $\mathbf{H}_N$ at the NMR frequencies. The degree of circular polarization was measured in a regime with separate counting of the right- and left-circularly polarized photons,[11] in analogy with the procedure described in our preceding paper.[7] The signals produced by the change of polarization $\Delta\rho$ under the NMR conditions were recorded also by the standard technique of synchronous detection at the oscillation frequency of a quartz phase plate that modulated the polarization in the luminescence-registration channel. The results cited below were obtained at 77 K. At liquid-helium temperature the picture of the phenomenon was qualitatively the same.

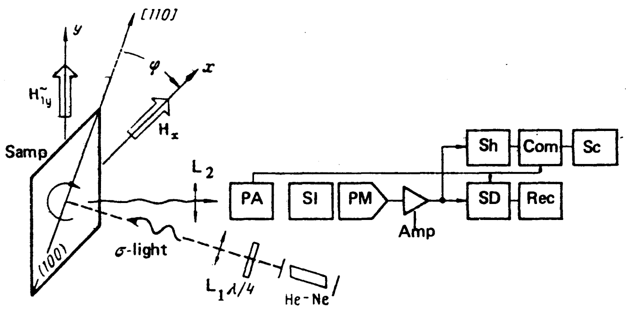

FIG. 1. Geometry of experiment. Samp—sample; L_1, L_2—lenses, λ/4—quarterwave plate, He-Ne—helium-neon laser, PA—quartz analyzer of luminescence polarization, SI—spectral instrument, PM—photomultiplier, Amp—narrow-band amplifier, SD—synchronous detector, Rec—automatic recorder, Sh—counting-pulse shaper, Com—pulse commutator, Sc—scaler setup.

2. Anisotropy of orientation of electron spins in a transverse magnetic field

Figure 2 shows a set of $\rho(H_x)$ plots obtained for different angles φ. As seen from the figure, a strong anisotropy is observed and is accompanied by qualitative changes of the function $\rho(H_x)$. A common feature of all these curves is the presence of a narrow peak near $H_x = 0$. With increasing field, the circular polarization changes strongly with changing angle φ.

A. Existence of two stable states of the spin-system polarization. At small φ ([110] axis close to the x axis) there exists a range of external fields where the polarization of the system of electron and nuclear spins has two stable states. On the boundaries of this range (H^b_{x1} and H^b_{x2} jumplike transitions take place between the two stable states. In the regions $H_x < H^b_{x1}$ and $H_x > H^b_{x2}$ the system of oriented spins can have only one stable state. The jumps of the circular polarization of the luminescence, which are observed in the experiment, are marked by the a-rows in Figs. 2a and 2b. When H_x increases from the weak-field side, a transition is observed at the point $H_x = H^b_{x2}$ from the state with higher polarization to a state with lower polarization. When H_x decreases from the strong-field side, the spin system remains in the state with the lower polarization down to the value $H_x = H^b_{x1}$. It is thus possible to realize any one of the two stable states in the range $H^b_{x1} < H_x < H^b_{x2}$.

The time of transition from one stable state to the other depends on the set values of the differences $\Delta H_{x1} = H^b_{x1} - H_x > 0$ and $\Delta H_{x2} = H_x - H^b_{x2} > 0$ for the lower and upper limits of the range with the two stable states, respectively. At $\Delta H_{x1,2} \sim 0$ the time of the transitions amounts to dozens of seconds. With increasing $\Delta H_{x1,2}$ this time decreases rapidly. Transitions between two stable states can be due not only to changes in the value of the field H_x. Such transitions can be obtained when the nuclear-spin system is acted upon by an alternating magnetic field or if the exciting light beam is turned off for a short time.

The boundary positions H^b_{x1} and H^b_{x2} are very sensitive to the orientation of the external field relative to the [110] axis. When the angle φ is varied in the range 0–3° the field H^b_{x2} changes by almost five times (from ~190 to ~40 Oe). The width $H^b_{x2} - H^b_{x1}$ of the region where the two stable states exist, changes correspondingly from 80 Oe to zero. Figures 2a and 2b illustrate the positions of the boundaries for two close values of the angle φ.

B. Slow oscillations of the luminescence polarization. An increase of the angle φ to a value at which the boundaries of the band with the two stable states come practically in contact lead to a qualitatively new phenomenon. A field region $H^{osc}_{x1} < H_x < H^{osc}_{x2}$ is produced, in which the polarization of the system of oriented spins executes slow undamped oscillations; near the boundaries, the oscillations are damped. This region is bounded by the dashed lines in Fig. 2c. With changing H_x, the amplitude and the period of the oscillations change. These data were cited in our preceding paper.[10]

Figure 3 shows the variation of the character of the

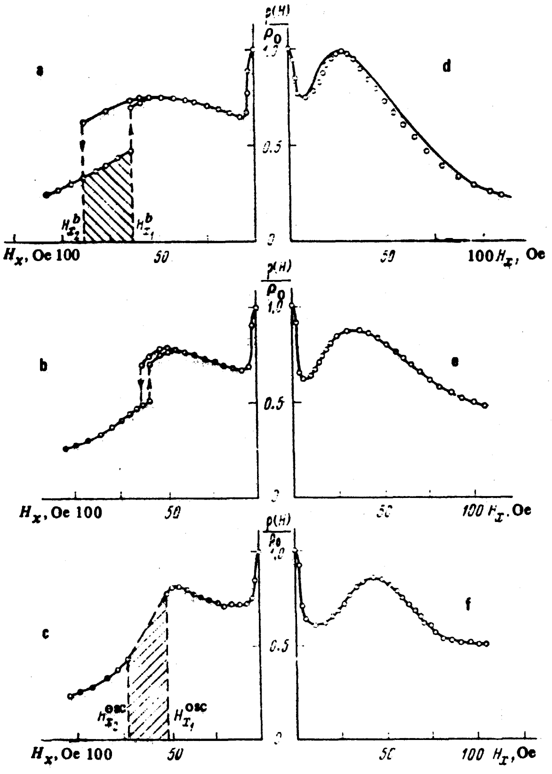

FIG. 2. Anisotropy of the $\rho(H_x)$ dependences (a) angle φ between the [110] axis and H_x equal to 1°; (b) $\varphi \approx 2,5°$; (c) $\varphi \approx 7,5°$; (d) $\varphi \approx 45°$ (the [100] axis coincides with H_x), the solid curve was calculated from formula (10) at $\alpha_e = 0.008$, $\beta = -18$, $H_L = 3.6$; (e) $\varphi \approx 35°$; (f) $\varphi \approx 18°$. All curves except (d) are drawn through the experimental points.

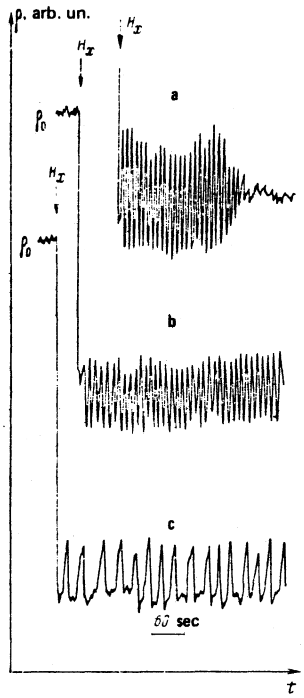

FIG. 3. Oscillations of the polarization luminescence, which are produced when a transverse magnetic field $H_x = 60$ Oe is applied, vs. the crystal orientation: (a) $\varphi \approx 5°$, (b) $\varphi \approx 7.5°$, (c) $\varphi \approx 12.5°$. The curves are drawn to the same scale but the origins are displaced. The arrows mark the instants when the field H_z is turned on.

oscillations with increasing angle φ. Near the boundary with the band of the hysteresis phenomena one observes oscillations (Fig. 3a) that are damped within a long time after the field has changed from $H_x = 0$ to a value of H_x within the band $H_{x2}^{osc} - H_{x1}^{osc}$ (the instant when H is turned on is marked by an arrow on Fig. 3). Further increase of the angle φ leads to undamped oscillations whose period is on the order of seconds and increases with increasing φ, as is illustrated in Figs. 3b and 3c. Figure 3 shows clearly the relaxation character of the oscillations.

The action of an alternating magnetic field at NMR frequencies can either stop the oscillations or change their period, depending on the values of the field H_x and of the angle φ. Conversely, in the field region $H_x > H_{x2}^b$ excitation of slow polarization is possible if an alternating magnetic field at the NMR frequency is applied.

If the direction of the magnetic field H_x is close to the [100] axis, no hysteresis effects are observed, and the oscillations shift into the region of much stronger fields. An experimental plot of $\rho(H_x)$ for $\varphi = 45°$ ($\mathbf{H}_x$ directed along [100]) is shown in Fig. 2d. It includes a narrow line near $H_z = 0$ and an additional maximum at $H_x \approx 30$ Oe. The value of ρ in the additional maximum approaches the value of ρ_0 at $H_x = 0$. The curve on Fig. 2d corresponds in all its details to the $\rho(H_x)$ function calculated by us on the basis of the model of cooling a spin system of nuclei in a field of optically oriented electrons.[7]

When the crystal is rotated around the z axis in a direction of decreasing φ, the minimum becomes deeper and the additional maximum drops lower (Figs. 2e, 2f).

Thus, strong anisotropy is produced in the case of optical orientation in a weak transverse field. It leads to various changes of the polarization in the optically oriented spin system of electrons and nuclei when the relative orientation of the external magnetic field and the crystallographic axes is changed. This shows that the model of cooling in the field of oriented electrons is inadequate and that it is necessary to take into account additional factors connected with the anisotropy of the hyperfine interaction. The necessary additional information on the structure of the hyperfine field will make it possible to observe the NMR spectra, which, as it turns out, can be easily detected optically even in a range of fields H that is so unusual for solid-state NMR spectroscopy.

2. Resonance effects connected with the anisotropy of nuclear polarization

A. Principal resonant transitions. The change of the field $\mathbf{H}_N$ at NMR frequencies under conditions when the derivative $d\langle S_z\rangle/dH_N$ is large enough makes it possible to detect nuclear magnetic resonance by an optical method. Optical detection of NMR via optical orientation of the nuclei in GaAlAs and GaAs crystals was effected previously in relatively strong fields (more than a kilooersted).[2,12,13] The change of the value of ρ was registered at frequencies γH, where γ is the nuclear gyromagnetic ratio. The dependence of $\langle S_z\rangle$ on the strong longitudinal external field and on the oblique external field was used.

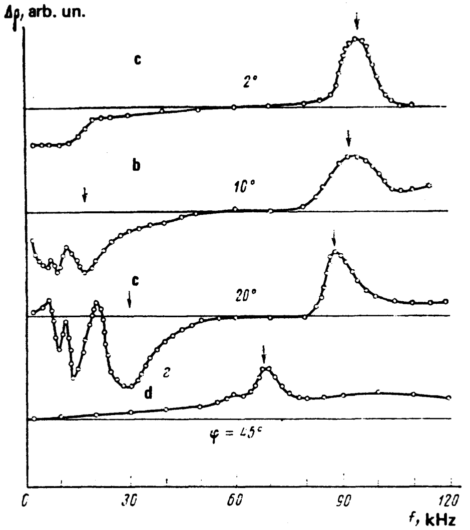

FIG. 4. NMR spectra at various orientation of the crystal in a field $H_x = 56$ Oe. Alternating field $H_{1y}^{\sim} = 2.8$ Oe. The arrows mark the positions of the principal resonances.

The high sensitivity of $\langle S_z\rangle$ to the value of $\mathbf{H}_N$ in the region of weak transverse fields makes it possible to register forbidden transitions and to obtain new information on the structure of the field $\mathbf{H}_N$. The substantial difference from the NMR spectra obtained in stronger external fields consists in the appearance of strong anisotropy.

Figure 4 shows the optically registered NMR spectra for different angles φ in a field $H_x = 56$ Oe. The spectra were obtained in the slow-passage regime. These spectra include two principal most intense lines, which come closer together when φ increases in the range from 0 to $\pi/4$, and then diverge as φ increases from $\pi/4$ to $\pi/2$. The behavior of these lines in a wide range of variation of the angles φ is described in a paper by Zakharchenya and the authors.[14] The angular dependences of the frequencies of the principal resonances consist of two branches that vary sinusoidally in the range between zero and $3K\gamma_i H$, where γ_i is the gyromagnetic ratio for the ^{75}As nuclei and $K \approx 0.82$.

Segments of the angular dependences for the upper (f_u) and lower (f_l) branches of the resonance frequencies are shown in Fig. 5a. We call attention to the sign of the resonance signal $\Delta\rho$. For the upper branch, the degree of circular polarization of the luminscence always increases. In the case of the lower branch the picture is more complicated. Depending on the angle φ and on the value of the constant field H_x and the alternating field $H_{1y}^{\sim}$ one can observe either positive or negative values of $\Delta\rho$. At large H_x ($H_x > 80$ Oe) and at relatively small $H_{1y}^{\sim}$ we have $\Delta\rho < 0$ for the lower branch. With decreasing H_x and increasing $H_{1y}^{\sim}$, signals where the zero level is crossed are observed, as shown in Fig. 4c. Further increase of the amplitude of the alternating field $H_{1y}^{\sim}$ is accompanied by an increase of the signal $\Delta\rho > 0$.

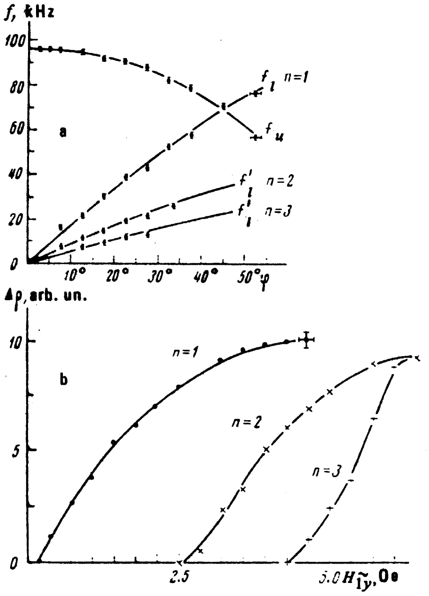

FIG. 5. Behavior of the principal resonances and of the subharmonic resonances at different angles φ and at different values of the alternating field $H_{1y}^{\sim}$: a—angular dependence of the resonance frequencies of the upper (f_u) and lower (f_l) branch with subharmonics (f_l'). b—dependence of the amplitude $\Delta\rho$ of the resonance signal on the alternating field $H_{1y}^{\sim}$ for the lower branch and the subharmonics. Field $H_x = 56$ Oe.

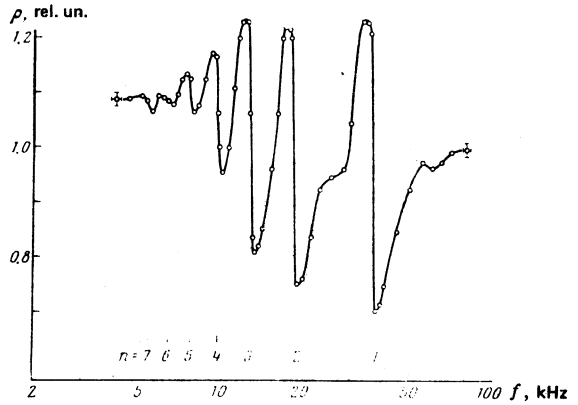

FIG. 6. Subharmonic resonances. The figure shows the section of the NMR spectrum corresponding to the lower frequency branch. $H_x = 56$ Oe, $H_{1y}^{\sim} = 6$ Oe, n is the number of the subharmonic. The level $\rho = 1$ corresponds to polarization at $H_{1y}^{\sim} = 0$.

At $\varphi = 0$ no resonances are observed on the lower branch. When the crystal is rotated into a position corresponding to $\varphi = 45°$, the amplitudes of the signals for both branches turn out to be comparable. Inasmuch as $f_u = f_l$ in this case, the resultant signal is weak (Fig. 4d). In the considered geometry, the maximum frequency ($\varphi = 0$) is $f_u \approx 0.82 \cdot 3\gamma_l H_x$. When the external magnetic field H_x is tilted towards the exciting-light beam, one can observe an additional approach of f_u to the value $3\gamma_l H_x$. The dependence of the frequencies f_u and f_l on the field H_x is close to linear. At H_x in the range 20–40 Oe, the picture becomes more complicated. In the region of f_l one observes an increase of $\Delta\rho$ when the frequencies vary in a range exceeding the widths of the principal resonances. The principal resonance at f_l is observed against the background of this general rise. Interesting singularities of the spectrum are observed in regions adjacent to the principal resonances on the low-frequency side.

B. Subharmonic resonances. The resonances observed at the frequencies $f_l' = f_l/n$, where $n = 2$ and $n = 3$, are well resolved on Fig. 4. By increasing the amplitude of the alternating field it is possible to separate lines with large n. Figure 6 shows the low-frequency section of the spectrum near the principal resonance of the lower branch at $H_{1y}^{\sim} = 6$ Oe. At a level exceeding three mean-square deviations one can clearly distinguish resonance transitions at subharmonics up to $n = 5$. For $n = 1, 2$, and 3 one observes abrupt changes of the resonance signal when the frequency is scanned from values for which $\Delta\rho < 0$ to values $\Delta\rho > 0$. The resonances at the subharmonics can be observed in a fixed range of angles φ and fields H_x. Figure 5 shows a summary of the results for the resonances at f_u, f_l, and f_l'; the experimental points on the $f_l'(\varphi)$ plots (Fig. 5a) give an idea of the range of angles φ in which it is possible to observe resonances on subharmonics at $n = 2$ and $n = 3$. Figure 5b illustrates the dependence of the subharmonic resonance signal $\Delta\rho$ on the field $H_{1y}^{\sim}$. It is seen that to obtain a noticeable NMR signal the value of $H_{1y}^{\sim}$ must exceed a certain threshold value. The threshold increases with the number of the subharmonic, and the regions of fields H_x in which subharmonic resonances can be observed decreases with increasing n. It should be noted that similar resonances, connected with the upper branch of the fundamental frequencies, are more weakly pronounced and are observed in a narrower interval of fields and angles.

C. Change of the state of the spin system in NMR. As indicated above, nuclear magnetic resonance can lead to changes of the state of a system of interacting nuclear and electron spins. NMR can give rise to transitions from one stable state to another, lead to excitation of slow undamped polarization oscillations, or cause the latter to be damped. By way of example, Fig. 7 shows transitions in a spin system at slow frequency scanning. We chose the angle φ and the field H_x such that the spin system has two stable states (Figs. 7a, 7b). By decreasing the field H_x below the boundary H_{x2}^b it is possible to establish a state with smaller polarization. If the alternating field $H_{1y}^{\sim}$ is small and the frequency f is scanned, one observes near f_l a resonance line (curve 1) similar to the one shown in Fig. 4. When $H_{1y}^{\sim}$ increases above the threshold value (≈ 1 Oe) the frequency scanning is accompanied by a jumplike transition from the lower to the upper stable state (curves 2 and 3 on Fig. 7b). These curves correspond to frequency scanning in opposite directions ($df/dt > 0$ for curve 3 and $df/dt < 0$ for curve 2). In both cases the polarization increases jumpwise at resonance, and the spin system remains subsequently in the upper stable state independently of the frequency of the alternating field. It is also possible to realize conditions under which the alternating field gives rise to a transition from the upper to the lower stable state.

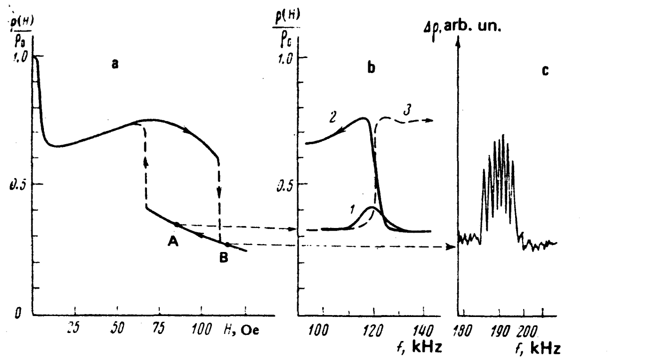

FIG. 7. Change of the state of the spin system when the alternating-field frequency is scanned near the NMR frequencies: a—plot of (H_x) with the field $\mathbf{H}_x$ oriented near the [110] axis, the arrows mark the direction of the change of the field H_x. The points A and B correspond to the values of the field H_x at which the action of the alternating field $H_{1y}^{\sim}$ was investigated; b—action of $H_{1y}^{\sim}$ on the spin system in the lower stables state (point A): (1) $H_{1y}^{\sim}=1.4$ Oe, (2, 3) $H_{1y}^{\sim}=3$ Oe; the arrows show the direction of the change of the frequency f; c—appearance of oscillations in passage of the resonance frequency in a strong field H_x (point B on Fig. a).

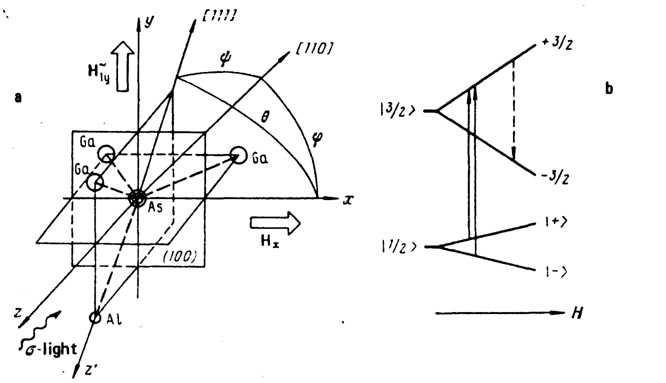

FIG. 8. Local disturbances of cubic symmetry when Al is substituted for one of the Ga atoms: a—the quadrupole interaction of the ^{75}As nucleus is determined by the gradient of the electric field along the Al—As bond (the z' axis); b—splitting of the energy levels of the ^{75}As nucleus in a weak magnetic field in the presence of a quadrupole interaction, the solid arrows show the scheme of the population of the level $+\frac{3}{2}$ when the semiconductor is excited by right-circular light, and the dashed lines correspond to the forbidden transition $+\frac{3}{2} \to -\frac{3}{2}$ in NMR.

Resonant transitions between two stable states are of interest, since they make it possible to enhance the resonance signal $\Delta\rho$ and to obtain a large value of $d\rho/df$. If $H_{1y}^{\sim}$ is chosen near the threshold value, the transition to the upper stable state occurs in the frequency band $\Delta f < 1$ kHz.

Figure 7c shows the onset of slow oscillations when the frequency f is scanned near f_u at a rate ≈ 0.15 kHz/sec. The field value is $H_x > H_{x2}^b$. It is interesting that in the absence of a field $H_{1y}^{\sim}$ in this band of angles φ and fields H_x no slow oscillations of the polarization are produced. We shall not dwell here in greater detail on the influence of the alternating-field amplitude or of the intensity of the exciting light, which affect strongly the shapes and number of the resolved resonance lines. The presented results assist in the choice of the optimal conditions for the observation of the maximum number of details in the spectrum.

3. DISCUSSION OF RESULTS

The aggregate of the presented results attests to the appearance of strong anisotropy of the hyperfine interaction between optically oriented electron and nuclear spins in solid semiconducting solutions. The model of cooling a spin system of nuclei in the field of optically oriented electrons[7] permits a complete description of the change of the circular polarization of the luminescence in a transverse field only when the [100] axis coincides in direction with the field. The strong changes in the form of the function $\rho(H_x)$ when the crystal is rotated demonstrate the need for taking into account additional factors connected with the crystal anisotropy. The resonance effects described above served as the key to the understanding of the nature of this anisotropy. The investigated crystals have a cubic lattice and their electrons have an isotropic g factor. At 77 K some of the nonequilibrium oriented electrons are situated in the conduction band and some are localized on shallow donor centers with a Bohr radius on the order of 100 Å. The total nuclear field $\mathbf{H}_N$, which acts on the electron spin, consists of the fields $\mathbf{H}_i$ of the individual isotopes, each of which can be in a different configuration:

$$\mathbf{H}_N = \sum_{i,r} P_r \mathbf{H}_i^{(r)}. \tag{1}$$

Here r denotes the number of the configuration that corresponds to a definite disposition of the neighboring nuclei.

We shall consider only the nearest-neighbor nuclei. If their total number is m and the probability of their replacing a nucleus of another species is x, then the quantity P_r is determined by the binomial distribution

$$P_r = \frac{m!}{r!(m-r)!} x^r (1-x)^{m-r}. \tag{2}$$

The number of the configuration is identified here only with the number of the substituted neighboring nuclei. It is assumed that there are no correlations between the substituting nuclei. For example, for ^{75}As $(m=4)$ in the solid solution $Al_xGa_{1-x}As$ with $x=0.24$ we have $P_0 \approx 0.315$; $P_1 \approx 0.42$; $P_2 \approx 0.21$; $P_3 \approx 0.05$ and $P_4 \approx 0.005$.

Thus, in most cases the nuclei ^{75}As either have no substituted neighboring nuclei, or one of the nearest neighboring Ga nucleus is replaced by Al. Since any of the Ga nuclei can be replaced, and the Ga–As bonds act along the body diagonals of the cubic cells, four directions turn out to be preferred. The cubic symmetry of the initial GaAs crystal turns out to be violated and local anisotropy sets in. It is obvious that one of the principal axes of the tensor of the electrical field gradient coincides with the direction to the substituted atom.

It is convenient to introduce local coordinate systems with axes z' along these directions (Fig. 8a). The ^{75}As nuclei have a quadrupole moment Q, and the Hamiltonian of the quadrupole interaction can be written, as usual, in the case of axial symmetry:

$$\hat{\mathscr{H}}=\frac{e^2qQ}{4I(2I-1)}[3\hat{I}_{z'}^2-I(I+1)]. \tag{3}$$

Here e is the electron charge, eq is the $z'z'$ component of the electric field gradient tensor, and $\hat{I}$ is the nuclear spin operator.

As will be made clear below, the quadrupole interaction that results from the substitution of Al for Ge, turns out to be such that observation of the forbidden transitions $\pm3/2\rightarrow\mp3/2$ becomes possible for ^{75}As nuclei in a weak magnetic field. A case is then realized wherein the nuclear Zeeman interaction can be regarded as a perturbation

$$\hat{\mathscr{H}}_{M}=-\hbar\gamma H\hat{I}_{z'}\cos\theta-{}^1/_2\hbar\gamma H\sin\theta(\hat{I}_+e^{-i\varphi'}+\hat{I}_-e^{i\varphi'}), \tag{4}$$

where θ and φ are the polar and azimuthal angles of the vector H in the local coordinate system.

In first-order perturbation theory, the splitting of the levels $\pm3/2$ is

$$hf=3\hbar\gamma H\cos\theta. \tag{5}$$

The observed two branches of the angular dependence of the resonance frequencies correspond to the contributions of two subgroups of the ^{75}As nuclei. The z' axes corresponding to different subgroups lie on mutually perpendicular planes, with $\cos\theta=\cos\psi\cos\varphi$ for the upper branch and $\cos\theta=\cos\psi\sin\varphi$ for the lower branch of the resonance frequencies. Here ψ is the angle between the z' axis and the [110] axis (see Fig. 9a). The experimentally measured values of the frequencies $f=3K\gamma H\cos\varphi$ correspond to (5), since the experimental value of K is close to the value $(2/3)^{1/2}$ of $\cos\psi$.

It can be shown that in third-order perturbation theory the splitting of the $\pm3/2$ levels is given by

$$hf=3\hbar\gamma H\cos\theta[1-2\alpha^2\sin^2\theta], \tag{6}$$

where $\alpha=\hbar\gamma H/e^2qQ$. The parameter α characterizes the relative values of the Zeeman and quadrupole energies. The deviation from the linear relation between f and H_x makes it possible to estimate α. This deviation could be reliably observed in the function $f_I(H_x)$ at small angles φ. In this case $\sin\theta$ is close to unity and the nonlinearity becomes more noticeable. The quantity α determined in the field $H_x=110$ Oe at $\varphi=25°$ amounted to 0.3 ± 0.1. This gives an idea of the scale of the quadrupole interaction.

The energy level scheme for nuclei with spin 3/2 in the case $\alpha\ll1$ corresponds to the one shown in Fig. 8b. The eigenstates $|+\rangle$ and $|-\rangle$ are mixed states defined by the expressions (see[15])

$$|+\rangle=|{}^1/_2\rangle\cos\delta+|-{}^1/_2\rangle\sin\delta,\quad |-\rangle=-|{}^1/_2\rangle\sin\delta+|-{}^1/_2\rangle\cos\delta.$$

Here δ is a function of the angle φ. In the regime of optical pumping with circularly polarized light, the oriented electrons lead, owing to the hyperfine interaction with the nuclei, to an increase of the population of either the +3/2 or −3/2 level, depending on the sign of the circular polarization of the exciting light. In this case transitions are allowed both from the levels $|+\rangle$ and from the levels $|-\rangle$, which correspond to superpositions of the states $|1/2\rangle$ and $|-1/2\rangle$. Direct relaxation of the optically produced population density is forbidden. The possible transitions are $\pm3/2\rightarrow|+\rangle\rightarrow\mp3/2$ or $\pm3/2\rightarrow|-\rangle\rightarrow\mp3/2$, and have a low probability.

Thus, optical pumping in the presence of quadrupole splitting leads to the appearance of a nuclear effective field component $P_1\mathbf{H}^{(1)}_{i,Q}$ in the sum (1). The alternating magnetic field $H_1^{\sim}$ equalizes the populations of the $\pm3/2$ levels and changes $\mathbf{H}^{(1)}_{i,Q}$, a fact that is registered in the optical interval by the change of the value of ρ. The probability $W_{(1)}$ of the one photon forbidden transition ($+3/2\rightarrow-3/2$) is proportional to the square of the modulus of the matrix element $\langle+3/2|H_1|-3/2\rangle$, where $\hat{H}_1$ is the energy operator of the perturbation by the alternating field. If the states $\langle+3/2|$ and $|-3/2\rangle$ are determined in first-order perturbation theory in the constant field, then this matrix element is equal to zero.

To find the probability $W_{(1)}$ it is necessary to determine four components of each of the state vectors $|+3/2\rangle$ and $|-3/2\rangle$, by solving the secular equation for the matrix $\mathcal{H}+\mathcal{H}_M$. Without dwelling on the details of the calculation, we present the final result:

$$W_{(1)}\approx(\gamma H_1^{\sim})^2\Gamma^{-1}\alpha^4\Psi(\theta,\xi). \tag{7}$$

Here Γ is the half-width of the resonances in hertz and $\Psi(\theta,\xi)$ is a function of the angle θ and of the angle ξ between the direction of the alternating field and the z' axis. In the calculation it is assumed that $H_1^{\sim}\ll H_x$ and $\mathbf{H}_1^{\sim}\perp\mathbf{H}_x$. The function $\Psi(\theta,\xi)$ varies in the considered range of angles from several units to several dozen.

At $H_1^{\sim}=5$ Oe, $\alpha=0.3$, and $\Gamma=10^4$ Hz, the order of magnitude of $W_{(1)}$ is 10^3–10^4 sec^{-1}, i.e., it is quite large. It is obvious that these resonant transitions can be observed in experiment provided that the 3/2-level pumping time is $T_{1e}\gtrsim1/W_{(1)}$. At the employed laser-beam intensities, the time T_{1e} ranged from several tenths of a second to several seconds. At these values T_{1e}, resonant changes of the field H_N can be observed also in relatively weak fields H_x (down to ~20 Oe), despite the fast decrease of $W_{(1)}$ with decreasing $H_x (W_{(1)}\sim H_x^4)$.

An interesting situation arises because of the absence an allowed transition and the possibility of observing a forbidden transition. The probability $W_{(1)}$ turns out to be comparable with the probability $W_{(2)}$ of two-photon transitions which are observed in suharmonic resonances (see Sec. 2, 3B). To estimate $W_{(2)}$ we consider the two-photon transition $+3/2\rightarrow-3/2$ via a virtual level that coincides with the level of the state $|+\rangle$. The probability of this transition is

$$\frac{4|\langle+{}^3/_2|\hat{\mathscr{H}}_1|+\rangle|^2|\langle+|\hat{\mathscr{H}}_1|-{}^3/_2\rangle|^2}{\Gamma(e^2qQ)}=\frac{9\hbar^2(\gamma H_1^{\sim})^4\sin^2\xi\,\mathrm{tg}^2\theta}{\Gamma(e^2qQ)^2(1+4\,\mathrm{tg}^2\theta)}. \tag{8}$$

Thus, we have in order of magnitude $W_{(2)}/W_{(1)}\sim(H_{1y}^{\sim})^2/H_x^2\alpha^2$.

It is easy to realize in experiment conditions under which the quantities $H_{1y}^{\sim}/H_x$ and α are of the same order.

The smallness of the two-photon transition can be characterized by the quantity α_1^2, where $\alpha_1 = \hbar\gamma H_{1y}^{\sim}/e^2qQ$. Obviously, the probability of three-photon transitions is $W_{(3)} \sim \alpha_1^4$. However, as shown by experiment, the resonance signal $\Delta\rho$ at $n=3$ can be comparable with the signals for $n=1$ and $n=2$. This situation is possible if $1/W_{(3)}$ remains less than T_{1e}. It should also be noted that the matrix elements that determine the probability of the three-photon transition do not contain small quantities in terms of the constant field, since this field includes three transitions that are allowed in the zeroth approximation. A more detailed analysis of the behavior of subharmonic resonances is beyond the scope of the present article.

The foregoing estimate of α shows that the frequency region of the pure quadrupole resonance (the transition $\pm 3/2 \to |+\rangle, |-\rangle$) can overlap the investigated frequency band. In fact, under certain pumping and crystal-orientation conditions one observes an appreciable increase of ρ in a relatively wide frequency range, which contains the estimated frequencies of the quadrupole resonance. Notice should be taken of the influence of the more remote substituted atoms on the frequencies of the quadrupole transitions, which can lead to the impossibility of resolving individual quadrupole-resonance lines. As to the transitions $\pm 3/2 \to \mp 3/2$, their energy remains practically unchanged when the quadrupole energy is changed, so long as the parameter α is small. Exact identification of the broad resonances calls for an additional analysis.

It is natural to raise the question of the reasons why the ^{75}As nuclei are singled out. Why do the analogous resonant transitions of the isotope nuclei ^{69}Ga and ^{71}Ga, which also have spin 3/2, manifest themselves weakly? We can cite the following reasons:

1) The number of nearest neighbors of the Ga nucleus, which can be replaced by Al, is equal to 12. The axial symmetry of the gradient of the electric field is then excluded already at small Al concentrations.

2) The As–Al distance is smaller by a factor $(3/8)^{1/2}$ than Ga–Al, and the quadrupole moment of the nuclei ^{69}Ga and ^{71}Ga is approximately half as large as that of ^{75}As. Other conditions being equal, these factors lead to a lowering of the energy of the quadrupole interaction of the Ga nuclei by almost one order of magnitude compared with the As nuclei.

We set aside the interesting question of the role of the ^{75}As nuclei with two neighboring Al nuclei, and consider ^{75}As nuclei that have no substituted neighboring nuclei. Together with ^{69}Ga, ^{71}Ga, and ^{27}Al they form a spin system that can be cooled to a reciprocal temperature $(\Theta_0)^{-1}$ [5]

$$\frac{1}{\Theta_0} \approx \frac{6\langle S\rangle H_x}{\mu(H_x^2+3H_L^2)} \tag{9}$$

μ is the averaged magnetic moment of the nuclei. This gives rise to a thermodynamic-equilibrium nuclear polarization along H_x and to a field $\mathbf{H}_{i,\Theta}^{(0)}$ at the electrons. The alternating field $H_{1y}^{\sim}$ "heats" the nuclear spin system and the field $H_{i,\Theta}^{(0)}$ decreases. It is impossible to separate in this case the usual resonant transitions with unity change of the magnetic quantum number for the individual isotopes. At large values of $H_{1y}^{\sim}$ one observes a variation of ρ in wide frequency ranges.[7]

NMR experiments point unequivocally to a large contribution of the states $|\pm 3/2\rangle$ of the ^{75}As to the formation of the field $\mathbf{H}_N$ under conditions of optical pumping in the presence of local anisotropy. The corresponding component of the field $\mathbf{H}_N$ can be represented in the form of the sum $P_1\mathbf{H}_{Q1}^{(1)} + P_1'\mathbf{H}_{Q2}^{(1)}$ (the symbol i has been omitted). The field $\mathbf{H}_{Q1}^{(1)}$ corresponds to the contribution of the quadrupole interaction for one pair of preferred directions (z_1', z_2'), while the field $\mathbf{H}_{Q2}^{(1)}$ corresponds to another pair (z_3', z_4'), so that $\mathbf{H}_{Q1}^{(1)}(\varphi) = \mathbf{H}_{Q2}^{(1)}(\varphi+\pi/2)$. For the [100] axis we have $\varphi = \pi/4$ and $|\mathbf{H}_{Q1}^{(1)}| = |\mathbf{H}_{Q2}^{(1)}|$. Attention should be called to the opposing actions of these fields. Under resonance conditions, by saturating the transitions $\pm 3/2 \to \mp 3/2$, we can alternately "erase" the fields $\mathbf{H}_{Q1}^{(1)}$ and $\mathbf{H}_{Q2}^{(1)}$, by using the frequencies f_l or f_u. As seen from Figs. 4 and 6, the polarization changes have in this case opposite signs.

Let us examine for the sake of argument the case of small φ (the [110] axis is close to the x axis). Let the field $H_{Q1}^{(1)}$ be turned off at the frequency f_u, and the field $H_{Q2}^{(1)}$ at the frequency f_l. At resonance with the frequency f_l, the quadrupoles for which the $z_3'z_4'$ plane is almost perpendicular to the x axis are turned off, and only the quadrupoles for which the $z_1'z_2'$ plane almost coincides with the xz plane are effective. The action of the external field is then enhanced on account of the field $\mathbf{H}_{Q2}^{(1)}$ and a negative signal $\Delta\rho$ is observed. At resonance at the frequency f_u, the field $H_{Q1}^{(1)}$ is turned off. The observed increase of ρ means then that in this case the action of the external field H_x is weakened. At $\varphi = 45°$ (symmetrical placement of H_x relative to the pairs of the quadrupole axes) the action of all the quadrupoles is cancelled out. The resultant resonant signal turns out to be small (see Fig. 4). In this case the field $\mathbf{H}_N$ is made up mainly of the fields $\mathbf{H}_i^{(0)}$.

The $\rho(H_x)$ curve shown in Fig. 2c is well described by an equation previously derived[7] on the basis of the model of cooling the spin system of nuclei in the field of optically oriented electrons

$$(1-\chi)(1+b)^2 = \alpha_e b\chi(1+b+\beta\chi)^2. \tag{10}$$

We have used here the dimensionless variables $\chi = \langle S_z\rangle/\langle S_z\rangle_0$ and $b = H_x^2/3H_L^2$, where $\langle S_z\rangle_0 = \rho$ at $H_x = 0$. The parameter β is the only parameter that characterizes the "depth" of the cooling. The quantity α_e determines the half-width of the $\rho(H_x)$ curve at $H_N = 0$. At the employed composition of the crystals, the g factor of the electrons is positive[16] and $\beta < 0$. The field $\mathbf{H}_{i,\Theta}^{(0)}$ is oriented opposite to the applied field. In the region of weak fields, where $H_N > H_x$, the depolarization is accelerated by the field $\mathbf{H}_N$ and a narrow line is observed. At $H_N \approx H_x$ the action of the external field is cancelled out and an additional maximum is produced. Further decrease takes place in the region where $H_x > H_N$.

If the condition $\alpha_e(|\beta|-1) > 4$ is satisfied, Eq. (9) has three solutions, of which one is unstable. The instability region can occur in the range of fields for which $\alpha_e b > 3$. It appears that the conditions realized experiments are close to these. The presence of the additional fields $H_{Q1}^{(1)}$ and $H_{Q2}^{(1)}$ contribute to the appearance of a region of multiply valued functions $\langle S_z \rangle$ of H_x, on the boundaries of which the polarization changes jumpwise. These very fields seem to facilitate the conditions for the onset of slow undamped polarization oscillations. These oscillations may be a reflection of the slow periodic changes of the reciprocal spin temperature $1/\Theta$.

D'yakonov and Perel' have shown[17] that the equation

$$\frac{d}{dt}\frac{1}{\Theta}=-\frac{1}{T_{1e}}\left(\frac{1}{\Theta}-\frac{1}{\Theta_0}\right)$$

describing the cooling of the spin system of the nuclei has at certain values of the parameters a unique stationary solution is unstable. The relaxation oscillations of Θ, and hence of the field H_N, lead to oscillations of the electron orientation, which is a function of Θ. The period of the oscillations is determined by the large time T_{1e}. The problem was solved for the case of optical orientation in a magnetic field parallel to the light beam. In the case of optical orientation in a transverse magnetic field it is more difficult to obtain the solution of the problem of the oscillations of the spin temperature. In addition, it is necessary to take into account in the initial equations the anisotropy. As follows from the experiment, the transitions $\pm 3/2 \rightarrow \mp 3/2$ influence strongly the state of the spin system. In NMR it is possible both to excite the oscillations and to cut them off, as well as to alter their waveform and the period. This points to the need for taking into account the influence of the quadrupoles when the conditions for the generation of slow oscillations are considered.

The aggregate of the described experiments demonstrates the strong manifestations of the local anisotropy of the semiconductor in optical orientation of electrons and nuclei in a relatively weak magnetic field. Cooling the nuclear spin system and anisotropy of the hyperfine interaction determine the peculiarities of the states, resonances, and behavior of an ensemble of interacting optically oriented electron and nuclear spins.

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Note added in proof (19 January 1978). A recent paper by V. L. Berkovits and V. I. Safarov {Pis'ma Zh. Eksp. Teor. Fiz. **26**, 377, 1977 [JETP Lett. **26**, 351 (1977)]} reports observation of resonances similar to those cited in Sec. 2.3.

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