Nonlinear effects in the electron-nuclear spin system of a semiconductor following absorption of light of variable circular polarization

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We investigate theoretically and experimentally nonlinear phenomena that accompany orientation of the electron-nuclear spin system of a semiconductor by light whose circular polarization oscillates in time at high frequency. We show that resonant absorption of the nuclear spin system in a field oscillating at close to the NMR frequency and produced by the oriented electrons at the nuclei can lead to the onset of bistable states of the electronic polarization. These states are connected with the partial cancellation of the external constant magnetic field by the effective nuclear field produced at the electrons by the polarized nuclei. We show in addition that a strong nuclear field can appear under nonresonant conditions if besides the modulation of the polarization of the exciting light one applies to the sample an external magnetic field that oscillates in phase with the average spin of the produced electrons. The appearance of this field is the cause of the instability of the electron-nuclear spin system in a zero constant magnetic field.

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§1. INTRODUCTION

Absorption of circularly polarized light produces in a semiconductors electrons whose average spin S_0 differs from zero. Owing to the hyperfine interaction, the crystal-lattice nuclei become polarized.[1] The polarized nuclei produce in turn an effective magnetic field $\mathbf{H}_N$ that acts on the electron spin.[2] The electron spins precess around $\mathbf{H}_N$. An interdependence of S and $\mathbf{H}_N$ sets in and leads to the onset of a strong feedback in the electron-nuclear spin system.

It was shown in Refs. 3–5 that when an electron-nuclear spin system becomes oriented by light of constant polarization, bistable polarization states and polarization instability can occur. These nonlinear effects were observed in $Al_xGa_{1-x}As$ solid solutions and were attributed to the nuclear-field anisotropy due to the quadrupole splitting of the spin levels of the arsenic nuclei when the Ga atoms were replaced by aluminum.[6]

It was noted in Ref. 7 that when the polarization of the exciting light is modulated at a sufficiently high frequency ω the field anisotropy vanishes. We shall show below that, nevertheless, the nonlinear effects mentioned above occur also in experiments with modulation. The point is that when the exciting light is modulated the resonant cooling of the nuclear system leads to the appearance of a strong nuclear field that depends on the spin of the oriented electrons and acts in turn on this spin. We shall show in §3 that the nuclear field can occur also under nonresonance conditions, when an external magnetic field $\mathbf{H}$ that oscillates in synchronism with $\mathbf{S}$ is applied to the sample simultaneously with the modulation of the exciting-light polarization. In this case, even if the constant magnetic field $\mathbf{H}_0$ is zero and $\mathbf{H}_1 \| \mathbf{S}_0$ (the external magnetic field does not depolarize the electrons) the nuclear field $\mathbf{H}_N$ turns out to be perpendicular to $\mathbf{S}_0$ and causes a decrease of the electron polarization.

§2. BISTABLE STATES OF RESONANTLY COOLED ELECTRON-NUCLEAR SPIN SYSTEM

As shown in Ref. 8, high-frequency modulation of the electron polarization $\mathbf{S}(t) = \mathbf{S}\cos\omega t$, $\omega T_2 \gg 1$, where T_2 is the characteristic time of the transverse relaxation of the nuclear spin) causes a noticeable polarization of the crystal-lattice nuclei only if the nuclear spins are simultaneously acted upon by a constant magnetic field $\mathbf{H}_0$ and by an alternating field $\mathbf{H}_1$ that oscillates[1)] at the same frequency ω as $\mathbf{S}$. The frequency ω_0 of the Larmor precession of the nuclear spins in the field $\mathbf{H}_0$ should then be close to ω (Resonant cooling). No account was taken in Ref. 8, however, of the strong reaction of the nuclear field on the polarization of the oriented electrons. We show below theoretically and experimentally that this reaction can lead to bistable states of the electron-nuclear spin system.

In the simplest case the value of the spin of oriented electrons located in a magnetic field $\mathbf{A}$ is determined by the equation

$$\mathbf{S}(t) - \mathbf{S}_0(t) = \hbar^{-1}\mu g T[\mathbf{A}\mathbf{S}(t)]. \tag{1}$$

Here $\mathbf{S}_0(t) = \mathbf{S}_0\cos\omega t$ is the instantaneous value of the average electron spin in the absence of the field $\mathbf{A}$, μ is the Bohr magneton, g is the electron g-factor, and T is the lifetime of the spin orientation. The electron spin will be given throughout in units of $\mathbf{S}_0$, and the unit of the magnetic field will be taken to be the half-width $H_{\frac{1}{2}} = \hbar/\mu g T$ of the electron-depolarization curve in a transverse magnetic field (of the Hanle curve). If the nuclei are polarized, the field $\mathbf{A}$ is a sum of the external magnetic field $\mathbf{H}$ and of the nuclear field $\mathbf{H}_N$:

$$\mathbf{A} = \mathbf{H} + \mathbf{H}_N, \tag{2}$$

and in the absence of crystal anisotropy the field $\mathbf{H}_N$ is parallel to $\mathbf{H}$ (Ref. 9). We shall be interested below in the case

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when the dc component of the external magnetic field is perpendicular to the propagation direction of the exciting light. If the spin of the oriented electrons oscillates at a sufficiently high frequency ($\omega T_2 \gg 1$), the field of the cooled nuclei is given by a general expression in the form

$$\mathbf{H}_N = h_N \langle \mathbf{I} \rangle = h_N h_e F(\Delta\omega) S^2 \mathbf{h}.$$

Here $\langle \mathbf{I} \rangle$ is the average spin of the polarized lattice nuclei, h_N and h_e are parameters that characterize the nuclear and electron fields ($\mathbf{H}_e = h_e \mathbf{S}$), $\mathbf{h}$ is a unit vector along $\mathbf{H}$, and the function $F(\Delta\omega)$ describes the cooling of the nuclear spin system and has a resonant dependence on the detuning $\Delta\omega = \omega - \omega_0$. Using (1), we easily find that $S_2 = \mathbf{S} \cdot \mathbf{S}_0 = S_z$ and

$$\mathbf{H}_N = h_N h_e F(\Delta\omega) S_z \mathbf{h}, \tag{3}$$

where S_z is the projection of the spin of the oriented electrons on the excitation direction. Substituting (2) in (1) and assuming that $\mathbf{S}_0 \perp \mathbf{H}$, we obtain a general equation for the z-projection of the electron spin in the case of cooling of a nuclear spin system in an oscillating electron field:

$$S_z = [1 + (h_N h_e F(\Delta\omega) S_z + H)^2]^{-1}. \tag{4}$$

It can be shown that at $H^2 < 3$ there is only one real value of S_z that satisfies (4). In the opposite case there can be three solutions.

A general analysis of the obtained cubic equation is quite cumbersome. If, however, the nuclear magnetic field is weak compared with H, ($h_N h_e F \ll 1$), Eq. (4) becomes linear

$$S_z = \frac{1}{1+H^2}\left(1 - \frac{2H}{(1+H^2)^2} h_N h_e F(\Delta\omega)\right). \tag{5}$$

In this simplest case to each value of H corresponds one value of the electron spin. In all the existing models, the dependence of F on $\Delta\omega$ is of the dispersion type. This leads to the appearance of characteristic singularities on the Hanle curve at resonant values of the magnetic field ($\omega_0 \approx \omega$).

If, however, $F h_N h_e \gtrsim 1$, it is easy to solve Eq. (4) numerically if the form of the function $F(\Delta\omega)$ is specified. The results of such a calculation for

$$h_N h_e F(\Delta\omega) = c \frac{\Delta\omega}{(\Delta\omega)^2 \omega_L^2} \tag{6}$$

(Ref. 8) are shown in Fig. 1. The parameters used in the calculations are: $c = -5.5$, $\omega_0 = 3$, $\omega_L^2 = 0.25$.

It can be seen from the calculated data that in a sufficiently strong magnetic field, when the Larmor-precession frequency ω_0 of the nuclear spin is close to the modulation frequency ω, there are three possible electron-polarization states. Large values of S_z corresponds to large values of the nuclear field that cancels out in part the external magnetic field H_0. (It it is of interest to note that a smooth transition into a state with high electron polarization as the magnetic field changes is impossible.) These states break away from the Hanle curve. When the magnetic field is smoothly varied, the dependence of S_z on H_0 can be described by the continuous line 1, and no highly polarized states appear. At the same time, if H is changed abruptly or some other action

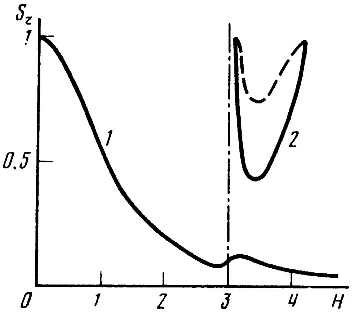

FIG. 1. Depolarization of electrons in a transverse magnetic field, with modulation of the circular polarization of the exciting light, as calculated with the aid of Eqs. (4) and (6) at $c = -5.5$, $\omega_0 = 3$, and $\omega_L^2 = 0.25$.

takes place the system can in principle go over into another stable state 2 that has a much larger electron polarization. This was in fact observed in experiment.

The measurements were performed with a setup similar to that described in Ref. 10. An $Al_{0.28}Ga_{0.72}As$ sample was excited with light from an He–Ne laser ($\epsilon_{h\nu} = 1.96$ eV) with alternating circular polarization. The polarization of the light was modulated with a piezoelastic quartz modulator at a frequency $\omega = 30.265$ kHz. The degree of polarization of the electrons was determined by measuring the degree ρ of the circular polarization of the edge radiation; in crystals such as GaAs this degree is numerically equal to the average spin of the oriented electrons $\langle S \rangle$. The value of ρ was determined by the method of two-channel counting of the left- and right-polarized photons in adjacent half-periods of the oscillations of the quartz modulator. The sample was cooled to the temperature of liquid helium. We measured the dependence of the projection S_z of the average electron spin on the direction of the exciting beam on the transverse magnetic field at $H = H_x$. The results of the experiment performed with an n-type $Al_{0.28}Ga_{0.72}As$ crystal at 4.2 K are shown in Fig. 2. It can be seen from this figure that bistability of the polarization is observed on the plot of the magnetic depolarization of the luminescence in the resonant-frequencies region. In some cases no states with high luminescence polar-

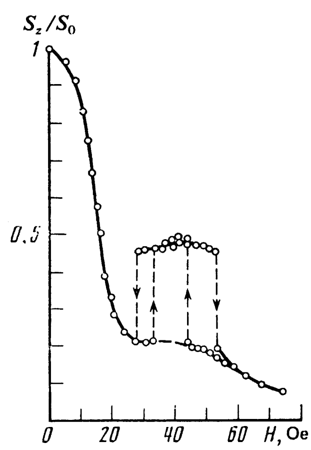

FIG. 2. Magnetic depolarization of electrons oriented by light in an n-type $Al_{0.28}Ga_{0.72}As$ crystal at 4.2 K and with modulation of the polarization of the exciting light at a frequency 30.265 kHz.

ization can be obtained by modulating only the polarization of the exciting light, in which case S_z decreases monotonically with increasing constant magnetic field. A transition into a stable state with high polarization can nevertheless be observed if the cooling action of the oscillating electric field is strengthened by applying an external alternating magnetic field $\mathbf{H}_1 \parallel \mathbf{S}_0$ in phase with the electron field. In this case the system remains in a state with high polarization after the field $\mathbf{H}_1$ is turned off.

The theoretical model developed above makes it possible thus to obtain qualitative agreement between the results of the calculations and the experiment. It becomes possible to understand the cause of the bistability and to determine the field region in which this effect is observed. It must be noted, however, that good quantitative agreement between experiment and calculation cannot be obtained. The disparity is apparently due to the simplified character of the model, which takes into account neither the inhomogeneities of the electric and nuclear fields, which appear when the electrons are localized on the impurity centers, nor the inhomogeneity of the distribution of the exciting-light intensity over the volume of the sample.

§3. INSTABILITY OF ELECTRON-NUCLEAR SPIN SYSTEM IN EXPERIMENTS WITH MODULATION IN THE ABSENCE OF A CONSTANT MAGNETIC FIELD

If the modulation frequency of the polarization of the oriented electrons is high, $\omega T_2 \gg 1$, then at $H_0 = 0$ the nuclear spin system is not cooled in an oscillating electric field.[11] Application of an alternating field $\mathbf{H}_1$ parallel to the direction of the exciting light and oscillating in phase with $\mathbf{S}_0$ should seemingly likewise not lead to cooling. In experiment, however, a strong decrease of the electron polarization is observed under these conditions.

Figure 3 shows the dependence of S_z on the amplitude of the alternating magnetic field H_1 in the absence of an external constant field. It can be seen from the figure that a weak alternating field exerts no noticeable influence on the electron polarization. When H_1 is increased above a certain critical value $H_{1\mathrm{cr}}$ the value of S_z begins to decrease, but with further increase of the field H_1 the average spin of the elec-

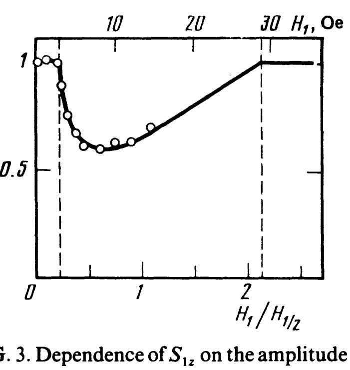

FIG. 3. Dependence of S_{1z} on the amplitude $\mathbf{H}_1$ of the alternating magnetic field parallel to $\mathbf{S}_0$ and oscillating in synchronism with $\mathbf{S}_0$ at a frequency 30.265 kHz. The region between the dashed lines corresponds to instability of the electron-nuclear spin system. The solid curve was calculated with the aid of (10) at $a = -5.6$ for $\varphi = +\pi$. The experimental points were obtained with an n-type $Al_{0.28}Ga_{0.72}As$ crystal at 4.2 K.

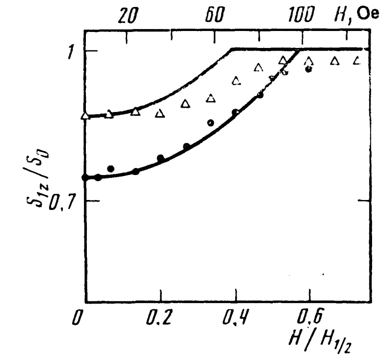

FIG. 4. Dependence of S_{1z} on the constant magnetic field $\mathbf{H}_0$ parallel to the exciting light beam. a) $\mathbf{H}_1 = 3$ Oe; b) $H_1 = 4.1$ Oe. The remaining conditions are the same as for Fig. 4. The solid curves were calculated with the aid of (11) for $a = -5.6$ at $\varphi = +\pi$.

trons increases again. An increase of the electron polarization at $H_1 > H_{1\mathrm{cr}}$ can be attained also by applying an external longitudinal constant magnetic field. These relations are plotted in Fig. 4 for two values of the field H_1.

The considered experimental results cannot be explained within the framework of the theory of cooling a nuclear spin system of a semiconductor by modulating the electron polarization.[11] The point is that even if the nuclear spin system were indeed cooled by the oriented electrons whose average spin oscillates at high frequency, this cooling could not produce in the absence of a constant magnetic field to a constant nuclear field that depolarizes the electrons, since the field of the cooled nuclei is directly proportional to H_0. In addition, it follows from the results of Ref. 11 that the cooling process itself is not very effective at such high modulation frequencies and at $H_0 = 0$.

We shall show that the experimentally observed decrease of the polarization can be due to the direct action of the alternating magnetic field on the spin of the oriented electrons, and the result is a noticeable nuclear polarization.

Indeed, if the average electronspin is not parallel to $\mathbf{H}_1$, the vector product $\mathbf{H}_1 \times \mathbf{S}$ in the right-hand side of (1) causes $\mathbf{S}(t)$ to contain a component that is constant in time even if the polarization of the exciting light is modulated at high frequency ($\omega T_2 \gg 1$). This dc component of the electron spin can lead to polarization of the lattice nuclei and to a nuclear field that depolarizes the electrons.[2)] It might seem that such a situation should be realized only if $\mathbf{H}_1$ is not parallel to $\mathbf{S}_0$. The theoretical calculation that follows, however, shows that at $\mathbf{H}_1 \| \mathbf{S}_0$ a state with high polarization of the electrons can be unstable. In this case another state can be stable, in which a large nuclear field perpendicular to $\mathbf{S}_0$ is produced.

We assume for simplicity that the alternating field H_1 is weak ($H_1 \ll 1$). The alternating (oscillating at the frequency ω) and the constant components of the electron spin are then determined by a system of equations that follows directly from (1):

$$\mathbf{S}_1 - \mathbf{S}_0 = [\mathbf{H}_N \times \mathbf{S}_1], \quad \langle \mathbf{S} \rangle - {}^1/_2 [\mathbf{H}_1 \times \mathbf{S}_1] \cos\varphi = [\mathbf{H}_N \times \mathbf{S}], \quad (7)$$

where $\mathbf{S}_1$ is the amplitude of the oscillating component of the electron spin. It is precisely this electron-spin component which is measured in experiment. $\langle \mathbf{S} \rangle$ is the time-averaged electron spin, φ is the phase shift between the exciting-light polarization oscillations and the alternating magnetic field ($\mathbf{S}_0(t) = \mathbf{S}_0 \cos \omega t$, $\mathbf{H}_i(t) = \mathbf{H}_i \cos(\omega t + \varphi)$. and the magnetic fields are measured, as before, in units of $H_{\frac{1}{2}} = \hbar/\mu g H$.

The nuclear-field $\mathbf{H}_N$ has a nonzero component at $H_0 = 0$ because of the polarization of the arsenic nuclei that undergo quadrupole splitting of the spin levels

$$\mathbf{H}_N = a\langle \mathbf{S} \rangle, \tag{8}$$

where the coefficient a does not depend on the external magnetic field.[9]

It can be easily seen that the system (7) and (8) has a trivial solution $\mathbf{S}_1 = \mathbf{S}_0$, $\langle \mathbf{S} \rangle = 0$. At a sufficiently large value of the parameter a which determines the value of the nuclear field, however, and at values of H_1 satisfying the condition

$${}^1/_2 H_1 a \cos \varphi \geqslant 1, \tag{9}$$

this solution turns out to be unstable. In this case another stable solution arises, for which the z-projection of $\mathbf{S}_1$ is given by

$$S_{1z} = ({}^1/_2 a H_1 \cos \varphi)^{-1}. \tag{10}$$

Corresponding to this solution are a nonzero component of the time-averaged spin $\langle \mathbf{S} \rangle \perp \mathbf{S}_0$ of the oriented electrons and a nuclear field $\mathbf{H}_N = a\langle \mathbf{S} \rangle$ perpendicular to $\mathbf{S}_0$, which causes the experimentally observed depolarization of the electrons.

The process that leads to the transition of the system from the unstable state with $\langle S \rangle = 0$ and $H_N = 0$ into the stable state (10) can be described in the following manner. Assume that a spin fluctuation was produced in the nuclear spin system, such that $\mathbf{H}'_N \perp \mathbf{S}_0$. Then $\mathbf{S}_1$ is no longer equal to $\mathbf{S}_0$ and contains a small increment $\mathbf{S}'_1 = \mathbf{H}'_N \times \mathbf{S}_0$. This causes a constant component $\langle \mathbf{S}' \rangle \sim [\mathbf{H}_1 \times [\mathbf{H}'_N \times \mathbf{S}_0]] \cos \varphi$ parallel to $\mathbf{H}'_N$. If the condition (9) is satisfied the onset of $\langle \mathbf{S}' \rangle$ leads to an increase of the initial value of the nuclear field. The fluctuation evolves until S_{1z} turns out to be equal to (10).

Figure 5 illustrates the onset of instability for antiphase modulation of $\mathbf{S}_0$ and $\mathbf{H}_1$ in the case $a < 0$. The flucutation of the field $\mathbf{H}'_N \perp \mathbf{S}_0$ causes rotation of the vector $\mathbf{S}_1$ in the xy plane. The deflection of $\mathbf{S}_1$ away from the z axis is accompanied by precession of this vector around $\mathbf{H}_1$. The oscillations of $\mathbf{H}_1$ in antiphase with $\mathbf{S}_1$ ensure the appearance of a dc component $\langle \mathbf{S} \rangle$ of the total average electron spin ($\mathbf{S} = \mathbf{S}_1 + \langle \mathbf{S} \rangle$), directed opposite to the fluctuation of $\mathbf{H}_N$ and amplifying this fluctuation, since $a < 0$.

Expression (10) was derived for the case $H_0 = 0$ and $H_1 \ll 1$. It can be generalized without difficulty to arbitrary values of the constant and alternating magnetic fields.

Thus, if $H_1 \ll 1$ but $\mathbf{H}_0 \neq 0$ and is parallel to $\mathbf{S}_0$, then

$$S_{1z} = 2(1 + H_0^2)/aH_1 \cos \varphi, \tag{11}$$

and the state with high polarization ($S = \mathbf{S}_0$) turns out to be unstable when

$$ {}^1/_2 a H_1 \cos \varphi (1 - H_0^2)/(1 + H_0^2)^2 > 1$$

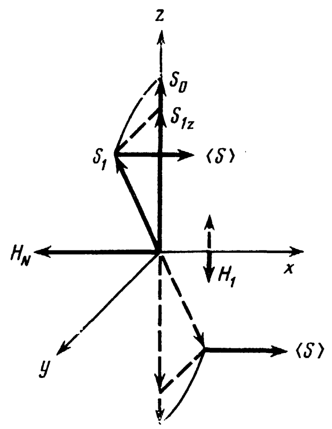

FIG. 5. Onset of instability of an electron-nuclear spin system under synchronous reorientation of the average electron spin $\mathbf{S}_0$ and of the field $\mathbf{H}_1$ parallel to it, in the absence of a constant magnetic field. The vectors in the upper half plane corresponds to the instant of time $t_1 = 2\pi n_i/\omega$, and in the lower half plane to the instants $t_i + \pi/\omega$.

It can be seen that the field H_0 stabilizes the state with $\mathbf{S}_1 = \mathbf{S}_0$.

Figure 4 shows the calculated dependence of the electron-spin projection S_{1z} on the longitudinal field H_0. It can be seen that when this field increases the polarization increases and at sufficiently large H_0 it reaches a value $S_{1z} = S_0$—the instability is eliminated.

A similar calculation can be carried out also for relatively large values of the alternating field ($H_1 \gtrsim 1$). In this case the final expression for S_{1z} is cumbersome and will not be presented here. We note only that strong alternating fields, just as a longitudinal field, stabilize the state $S_{1z} = S_0$. The results of the corresponding calculation are shown in Fig. 3.

Experiment reveals all the indicated distinguishing features: (1) the presence of a threshold value H_1 starting with which the electron polarization in a zero constant magnetic field decreases, (2) the onset of instability when $\mathbf{S}_0$ and $\mathbf{H}_1$ oscillate in antiphase and the absence of a decrease of the polarization when $\mathbf{S}_0$ and $\mathbf{H}_1$ oscillate in phase (at $a < 0$), (3) stabilizing action of a constant magnetic field $\mathbf{H}_0 \| \mathbf{S}_0$ and of a strong alternating field $\mathbf{H}_1$.

The calculated curves are in satisfactory quantitative agreement with the measurements results. This allows us to conclude that the considered simple model of the instability in a zero magnetic field describes the real situation sufficiently well.

CONCLUSION

The presented results of the experimental and theoretical investigations show that under conditions of high-frequency modulation of the polarization of the exciting light ($\omega T_2 \gg 1$) the orientation of the electrons can lead to a considerable polarization of the crystal-lattice nuclei. As a result, just as in constant pumping, a strongly coupled electron-nuclear spin system is produced with its inherent salient features (bistable states and instability). If the nuclear spin system is cooled in an oscillating field of oriented electrons, a strong feedback between the nuclear and electron spins is pronounced when the modulation frequency is close to the resonant precession frequency of the nuclear spin in a constant magnetic field (resonant cooling). If an external magnetic field oscillating at the same frequency ω is applied to the sample, the onset of the nuclear field may be connected with the appearance of a time-constant component of the average spin of the oriented electrons. This effect takes place even in the absence of a constant magnetic field and at parallel $\mathbf{S}_0$ and $\mathbf{H}_1$.

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[1)] The role of the alternating field H_1 can be played by the electron field H_e produced at the nuclei by the oriented electrons via the hyperfine interaction. This is precisely the case analyzed in the present section.

[2)] As shown in Ref. 12, in the $Al_xGa_{1-x}As$ crystal on which the experiment described above was performed, the quadrupole splitting of the arsenic-nuclei spin levels causes the time-constant component of the electric field to produce a noticeable nuclear polarization even at $H_0 = 0$.

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