

B. P. Zakharchenya, V. A. Novikov, and V. G. Fleisher, *States and resonances of optically pumped electron and nuclear spins in a semiconductor*. Optical pumping makes it possible to achieve a high degree of orientation of electron and nuclear spins by simple means. Recent research on semiconductors has established several basic aspects of the behavior of optically pumped spin systems in solids. In solids, in contrast with gases, the strong spin-spin interactions in the nuclear system are important, and they lead to a spin temperature which is different from the lattice temperature. Furthermore, it turns out that the behavior of the electron spins in weak external magnetic fields is affected by an anisotropy of the hyperfine field and, in particular, nuclear quadrupole effects.

Study of the anisotropy of optical pumping has led to the discovery of fixed stable states of electron-nucleus spin systems, with both constant and continuously oscillating polarizations. Transitions between these states can be caused by various external factors, in particular, alternating magnetic fields at NMR frequencies. In the optical channel, magnetic resonances are easily detected in weak fields and low frequencies, which are unusual for solids. These resonances make it possible to clearly identify the contribution of nuclear quadrupole effects.

Let us examine some results obtained in a study of optical orientation in the semiconducting solid solution $\text{Ga}_x\text{Al}_{1-x}\text{As}$ in which $\sim 1/4$ of the Ga atoms of the original GaAs crystal are replaced by Al atoms ($x=0.24$). These *p*-type crystals (doped with Zn to a concentration $\sim 10^{18} \text{ cm}^{-3}$) were excited by the beam from a He-Ne laser, which was incident normally on the crystal surface along the fourfold axis. Most of the measurements were carried out in an external magnetic field H directed perpendicular to the light beam. The crystallographic axes were in various orientations with respect to this field; the orientation was changed by rotating the crystal around the laser beam. In the experiments, measurements were made of the quantity ρ , which is the degree of circular polarization of the luminescence. This quantity is numerically equal to $SS_1/|S_1|$, the projection of the average electron spin S onto the propagation direction of the exciting light. (Here $|S_1| = \rho$ is the average spin of the electrons which are oriented by the light in a zero magnetic field.) The measurements were carried out at 77°K. The depolarization of the luminescence in a transverse magnetic field, which is sometimes called the "Hanle effect" by analogy with gases, can yield much information on the hyperfine field in the case of semiconductors. The electrons oriented by the light cool the nuclear spin system to a reciprocal temperature $[1/\Theta \sim (SH)/(H^2 + H_{10c}^2)]^{1,2}$ (H_{10c} is the local nuclear field) by virtue of the hyperfine interaction.^{1,2} In the geometric

arrangement under consideration, this cooling is possible in the field $h_e S$ of the electrons oriented by the light^{3,4} ($h_e = \text{const}$). In this case, $[1/\Theta \sim S^2(H \gg h_e S)]$. When thermodynamic equilibrium is reached, the light-cooled nuclear spin system is polarized in the direction of the external field H . Since the light causes a substantial cooling, even in a weak external field (of a few tens of oersteds), a substantial nuclear polarization can result. The polarized nuclei create an effective magnetic field H_N , which acts on the electron spins. In weak fields H this effective field can accelerate the luminescence depolarization, leading to the appearance of a characteristic narrow line in a plot of $\rho(H)$ (Refs. 3 and 4). If $H_N \uparrow \uparrow H$, there is a region in which the external field is approximately cancelled by the internal field, and the quantity ρ turns out to be essentially the same as in the case $H=0$. An additional rising region appears on the $\rho(H)$ curve. The solution of the Bloch equation describing the behavior of the electron spin S in a transverse field, with cooling of the nuclear spin system, leads to a good agreement with experiment in those cases in which the anisotropy is not important. On the basis of this model it was predicted that there are two stable polarization states near the region in which the external and internal fields cancel out. Soon after this prediction was made, these states were in fact detected experimentally.⁵ At the boundaries of the range of H in which the function $\rho(H)$ is multivalued the luminescence polarization changes abruptly. The size of this range determines the width of the "hysteresis loop."

The function $\rho(H)$ can change markedly when there is a change in the orientation of the crystal. A rotation of the crystal around the laser beam can cause transitions from states with a constant polarization to states with a continuously oscillating polarization. The electron-nuclear spin system turns out to be unstable under constant external conditions. Since the electron *g*-factor is isotropic in the cubic crystals involved here, the only anisotropy is that due to the hyperfine field. Optical detection of nuclear magnetic resonances has made it possible to identify the nature of this anisotropy.^{6,7} Figure 1 shows the resonant frequencies f of the principal lines in the NMR spectrum as functions of φ , the angle between the $\{110\}$ axis and the external field H . The resonances were inferred from the change in the quantity ρ upon the imposition of an alternating field $H_N \perp S_1$, H . A change in the effective field H_N at resonance causes a change in ρ . As Fig. 1 shows, all the resonant frequencies conform well to two sinusoidal branches. The absolute values of the frequencies correspond to three times the frequency of the ordinary As⁷⁵ nuclear magnetic resonance in magnetic fields which are equal to the projections of the external field onto the body diagonals of the cube. In this geometric arrangement, these pro-

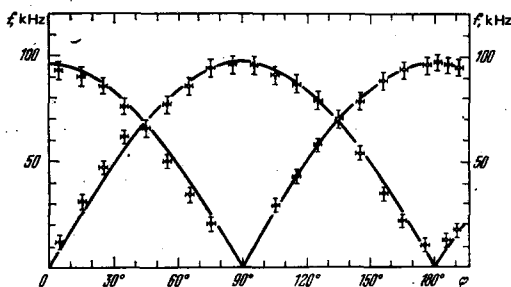


FIG. 1. Angular dependence of the resonance frequencies.

jections differ in pairs for the four principal diagonals, so that there are two frequency branches. When the field H makes some angle with respect to the laser beam, each branch again splits in two.

The observed pattern of nuclear resonances corresponds to the quadrupole interaction which results from a local deviation from symmetry when some of the Ga atoms are replaced by Al atoms. The appearance of electric field gradients at the As nuclei nearest the substituted atoms leads to a quadrupole splitting of the $1/2$ and $3/2$ levels. The external magnetic field splits the $3/2$ levels by an amount $3H\cos\psi\cos\varphi$ for one pair of quadrupoles and $3H\cos\psi\sin\varphi$ for the other pair. Here ψ is the angle between the body diagonal of the cube and the face diagonal. With the optimum orientation, either the $+3/2$ or $-3/2$ levels are populated preferentially, depending on the sign of the circular polarization of the light. An additional component of the field H_N arises and affects the value of ρ . When the populations of the $\pm 3/2$ levels are made equal under NMR conditions, it is a simple matter to detect in the optical channel those effects which are due to the nuclear quadrupole interaction.

A change in the magnitude of the field H_N under resonant conditions can lead to qualitative changes in the states of the spin system. Figure 2 shows those changes which occur as the frequency of the alternating field is slowly varied. Figure 2a shows the behavior $\rho(H)/\rho_1$ for $\varphi=0$. This plot includes a region in which there are two stable polarization states. At the boundaries of this region, the luminescence polarization changes abruptly, as shown by the arrows in Fig. 2a. We thus have an optical analog of a switching effect. Transitions between the stable states can occur not only at the boundaries of the hysteresis region but also inside it. For this purpose it is sufficient to change the field H_N , for example, with the aid of an alternating field H at the NMR frequency. Curves 1 and 2 in Fig. 2b were found by varying the frequency of H near the resonance at a rate ~ 0.15 kHz/sec. The polarization changes from the value corresponding to the lower stable state at the point A in Fig. 2a to the values corresponding to the upper stable state.

Figure 2c demonstrates the appearance of slow polar-

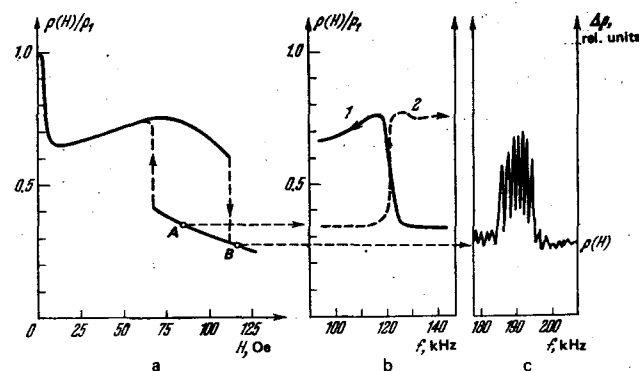


FIG. 2. Change in the state of the spin system as the frequency of the alternating field is varied. a) $\rho(H)/\rho_1$ for $\varphi=0$ and $H_N=0$; b) transition from the lower stable state to the upper one for point A in Fig. 2a; c) onset of oscillations with a period ~ 10 sec for point B in Fig. 2a.

ization oscillations with a period ~ 10 sec as the frequency of the alternating field is varied at the same rate, but with the field H held constant at a value corresponding to point B in Fig. 1a. In the absence of the field H , this point corresponds to the sole stable state with a constant polarization. If the frequency scanning is stopped in the resonant region, the luminescence polarization undergoes undamped oscillations. Such oscillations can be observed even in the absence of an alternating field,⁵ if the angle φ is increased to 5° – 10° .

The strong mutual effects of the electron and nuclear polarizations during optical orientation of spins thus lead to several new magneto-optical effects, which appear in weak fields (an unusual situation for the magneto-optics of semiconductors). The cooling of the nuclear spin system and the anisotropy of the hyperfine interaction determine the distinctive nature of the states and resonances of an ensemble of optically pumped electron and nuclear spins.

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Translated by Dave Parsons