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OPTICAL DETECTION OF DYNAMIC POLARIZATION OF NUCLEI IN SEMICONDUCTORS

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Much research has been performed by now on optical pumping of spin-polarized electrons in semiconductors [1 - 6]. In these experiments, the spin-oriented carriers were excited from the valence band into the conduction band by absorption of circularly-polarized light. The most convenient for the detection of the spin orientation of the electrons is an optical method [2, 3] with investigation of the recombination-luminescence polarization. For III-V crystals, the selection rules for interband transitions yield a simple connection $S = 0.5P$ between the degree of circular polarization of the luminescence (S) and the degree of stationary orientation of the electrons (P) at the bottom of the conduction band. We have investigated, also by an optical method, a number of effects that arise when the electron spins interact strongly with the magnetic moments of the nuclei in the crystal.

As is well known, electron-nucleus collisions can be accompanied by mutual spin flip. In the case when the electrons are strongly polarized, the electron orientation becomes transferred to the nuclei by such collisions, i.e., dynamic polarization of the nuclei takes place. Such a phenomenon was first observed by Lampel [1] and revealed by the increase of the nuclear-magnetic resonance signal from a silicon crystal illuminated by circularly polarized light. Such a dynamic polarization of nuclei can be revealed also by the change of the degree of stationary orientation of the electrons, i.e., by optical means (by the luminescence polarization).

Let us examine the differential equations describing the relaxation processes of oriented electrons in a crystal (for simplicity we assume that the magnetic moments of the nuclei are equal to $\pm 1/2$):

$$\frac{dN_{\pm}}{dt} = I G_{\pm} - \frac{N_{\pm}}{T} - \frac{N_{\pm} - N_{\mp}}{2T_s} - (N_{\pm}n_{\mp} - N_{\mp}n_{\pm}) \frac{W}{2} \quad (1)$$

$$\frac{dn_{\pm}}{dt} = - \frac{n_{\pm} - n_{\mp}}{2\tau_s} + (N_{\pm}n_{\mp} - N_{\mp}n_{\pm}) \frac{W}{2}$$

Here N_{\pm} and n_{\pm} are the densities of the electrons and nuclei with the corresponding spins, I is the intensity of the exciting light, G the probability of producing an electron with different spin orientation upon absorption of circularly polarized light, T the lifetime of the nonequilibrium electron, and T_s and τ_s the spin-lattice relaxation times of the electrons and the nuclei. The last terms in the equations take into account the mutual spin flips of the electrons and of the nucleus, and W is the probability of such a flip upon collision. The solution of the equations in the stationary case yields

$$P = P_0 \left[1 + \frac{T}{T_s} + \frac{T}{T_n} (1 - f) \right]^{-1} \quad (2)$$

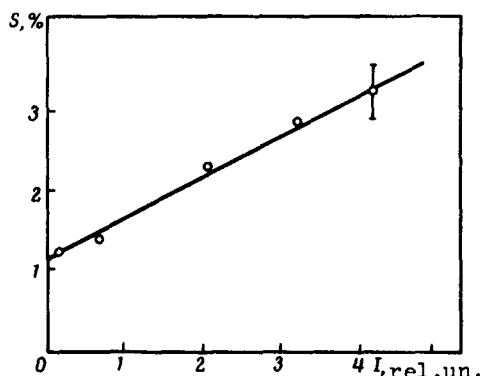


Fig. 1. Degree of luminescence polarization of a p-type $\text{Ga}_{0.7}\text{Al}_{0.3}\text{As}$ crystal vs. excitation intensity at 4.2°K .

trons collide with the nuclei so infrequently that there is enough time between two collisions for the nuclei to lose the orientation transferred to them as a result, say, of spin-lattice relaxation ($f \rightarrow 0$). The electrons will therefore constantly collide with unpolarized nuclei. At high excitation intensity, however, the collisions can occur so frequently that the nucleus retains its orientation in the time between two collisions ($f \rightarrow 1$). Therefore the spin relaxation of the electrons on the nuclei slows down, causing an increase in the degree of stationary orientation. If $T \approx T_s \gg T_n$, the P depends linearly on the light intensity, and as $I \rightarrow 0$ it tends to a certain value determined by the relaxation of the electrons on the unpolarized nuclei.

Such a phenomenon was observed by us in p- $\text{Ga}_{0.7}\text{Al}_{0.3}\text{As}$ crystals, in which all the atoms of the principal lattice have nuclear magnetic moments. The excitation was with an He-Ne laser. Figure 1 shows the degree of the circular polarization of the crystal luminescence (S) at 4.2°K as a function of the intensity of the circularly polarized exciting light (I). We see that an increase of I is accompanied by a linear increase of the degree of polarization from approximately 1 to 3%.

Interesting phenomena should be observed in an external magnetic field. In the usual case, an external magnetic field perpendicular to the orientation of the electron spins depolarizes the electrons (the Hanle effect) as a result of the precession of the spins about the direction of the magnetic field, which occurs at a frequency $\Omega = e\hbar H/2mc$. The $P(H)$ plot has in this case a Lorentz form, $P(H) = P(0) [1 + \Omega^2 T_0^2]^{-1}$, and its half-width is determined by the condition $\Omega T_0 = 1$, where $1/T_0 = 1/T + 1T_s$. In our case, solution of the corresponding equations gives a more complicated relation

$$P(H) = \frac{P_0 \left[1 + \frac{T}{T_s} + \frac{T}{T_n} \left(1 - \frac{f}{\omega^2 \tau^2 + 1} \right) \right]^{-1}}{\left[\Omega + \frac{\omega \tau f}{T_n (\omega^2 \tau^2 + 1)} \right]^2 \left[\frac{1}{T} + \frac{1}{T_s} + \frac{1}{T_n} \left(1 - \frac{f}{\omega^2 \tau^2 + 1} \right) \right]^{-2} + 1}, \quad (3)$$

where ω is the nuclear-spin precession frequency (usually $\omega \ll \Omega$) and $1/\tau = 1/\tau_e + 1/\tau_s$.

Here P_0 is determined by the values of G_{\pm} ($P_0 = 0.5$ for $\Gamma_{8v} - \Gamma_{6c}$ transitions in III-V crystals), f is the polarization factor of the nuclear spins, $f = \tau_s / (\tau_e + \tau_s)$, where $T_n = (Wn/2)^{-1}$ and $\tau_e = (WN/2)^{-1}$ are none other than the times for the spin relaxation of the electrons on the nuclei and conversely (N and n are the total numbers of nonequilibrium electrons and nuclei). Since N depends on the intensity of the exciting light ($N = iT$), the polarization factor f and the degree of stationary orientation of the electrons P also depend on the excitation intensity.

This result can be explained as follows. At low oriented-electron concentration (low excitation intensity) the elec-

trons collide with the nuclei so infrequently that there is enough time between two collisions for the nuclei to lose the orientation transferred to them as a result, say, of spin-lattice relaxation ($f \rightarrow 0$).

The electrons will therefore constantly collide with unpolarized nuclei. At high excitation intensity, however, the collisions can occur so frequently that the nucleus retains its orientation in the time between two collisions ($f \rightarrow 1$). Therefore the spin relaxation of the electrons on the nuclei slows down, causing an increase in the degree of stationary orientation. If $T \approx T_s \gg T_n$, the P depends linearly on the light intensity, and as $I \rightarrow 0$ it tends to a certain value determined by the relaxation of the electrons on the unpolarized nuclei.

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where ω is the nuclear-spin precession frequency (usually $\omega \ll \Omega$) and $1/\tau = 1/\tau_e + 1/\tau_s$.

In the case of low excitation intensity ($f = 0$), this formula reduces to the customary one for the Hanle effect, and the half-width is determined by the relaxation of the electrons on the unpolarized nuclei. At high light intensity ($f \neq 0$), an important role is played by the nuclear polarization but, as seen from (3), at $H \neq 0$ the nuclear polarization factor must be replaced by the expression $f/(\omega^2\tau^2 + 1)$. With increasing H , this expression tends rapidly to zero (the term $\omega^2\tau^2$ increases rapidly since the nuclear spin relaxation times are very large). Thus, even at small magnetic-field intensities (when we still have $\Omega T_0 \ll 1$), the degree of electron orientation decreases to a value corresponding to a low light intensity ($f/(\omega^2\tau^2 + 1) \rightarrow 0$). This is attributable to the fact that the external field, by causing precession of the nuclear spins, depolarizes thereby the nuclei; under such conditions, the degree of electron orientation is determined already by relaxation on non-oriented nuclei. With further increase of the magnetic field, the observed Lorentz contour will be the same (will have the same amplitude and half-width) as in the case of low light intensity.

Figure 2 shows the obtained dependence of the degree of luminescence polarization on the transverse-field intensity in the investigated $\text{Ga}_{0.7}\text{Al}_{0.3}\text{As}$ crystal for two values of the excitation intensity. We see that the observed dependences correspond fully to the model under consideration. The fall-off of the degree of polarization at low field intensities (large excitation intensities) constitutes in essence the Hanle effect for the polarized nuclei. From the obtained $P(H)$ curve we can determine the spin relaxation times of the electrons ($T_n \approx 10^{-9}$ sec) and of the nuclei ($\tau \approx 10^{-3}$ sec).

We note that in the presence of a longitudinal magnetic field the nuclei can be depolarized by applying a radio-frequency field (saturation of the magnetic resonance). We have observed a decrease in the degree of luminescence polarization in the investigated crystals upon application of a radio-frequency field of 180 kHz (longitudinal dc field $H = 145$ G), which corresponds closely to resonance with the Ga nuclei.

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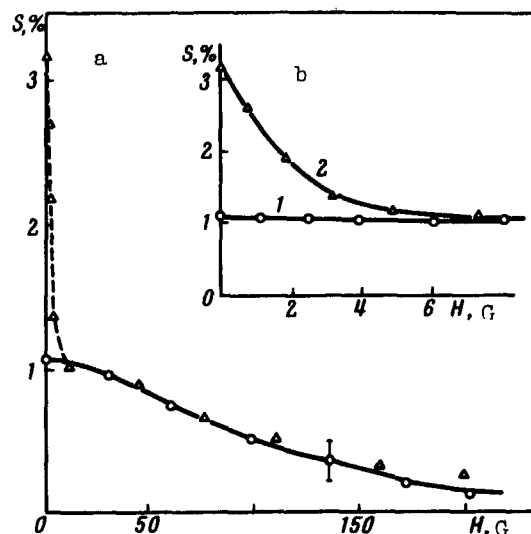


Fig. 2. Degree of luminescence polarization of $\text{Ga}_{0.7}\text{Al}_{0.3}\text{As}$ crystals vs. transverse magnetic field intensity, for different excitation intensities: 1 - excitation weakened by a factor of 10, 2 - maximum excitation. The solid curve in a is a Lorentz contour with half-width 110 G.