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## Instabilities in the spin system of optically oriented electrons and nuclei in semiconductors

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It is shown that quadrupole splitting of the spin levels of the arsenic nuclei in  $Ga_xAl_{1-x}As$  solid solutions makes possible slow rotation of the field of the dynamically polarized nuclei. These rotations are connected not with the rotation of individual nuclear spins, but with the change in the intensity of the field produced by the nuclei for which the quadrupole-interaction axes are differently oriented. It is shown that this leads, under certain conditions, to instability of the stationary states in the system of optically oriented electrons and nuclei of the semiconductor and to the onset of oscillations of the degree of polarization of the recombination radiation, with a period determined by the time of the longitudinal spin relaxation of the lattice nuclei. The results are compared with the experimental data.

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### 1. INTRODUCTION

Interband absorption of circularly polarized light in a semiconductor is accompanied by spin orientation of the photoexcited electrons, which leads to dynamic polarization of the crystal-lattice nuclei. The polarized nuclei act in turn on the electron orientation via the effective magnetic field produced by the hyperfine interaction.<sup>1</sup> Thus, a nonlinear system of coupled electron and nuclear spins is produced. The nonlinearity of this system manifests itself, for example, in hysteresis of the dependence of the degree of polarization of the recombination radiation on the external magnetic field.<sup>2,3</sup> A number of studies have revealed phenomena that attest to the onset of instabilities in this system<sup>4</sup>

and to the onset of associated self-oscillations.<sup>3,5</sup> These experiments were performed on  $Ga_xAl_{1-x}As$  solid solutions, in which an essential role is played by the quadrupole splittings of the nuclear spin levels, due to local disturbances of the cubic symmetry.<sup>2,3,5-8</sup>

The quadrupole splitting in  $Ga_xAl_{1-x}As$  solid solutions is particularly substantial for the arsenic nuclei, for which the cubic symmetry of the environment is violated even in the first coordination sphere if one or several gallium atoms in this sphere are replaced by aluminum atoms. This is confirmed by experiments on optically detected nuclear magnetic resonance.<sup>3,5,6</sup> The quadrupole interactions manifest themselves, in particular, in the strong dependence of the shape of the Hanle curve on the direction of the external magnetic

A theoretical analysis of the optical orientation in the system of electron and nuclear spins for stationary conditions was carried out in our preceding paper.<sup>10</sup> The present paper is devoted to a study of the instabilities and self-oscillations in this system. We shall show that these effects can be due to the quadrupole splittings of the nuclear spin levels that takes place in the solid solutions.

## 2. MODEL AND BASIC EQUATIONS

It was shown in Ref. 10 that the experimentally observed Hanle curves are qualitatively described correctly on the basis of the general symmetry properties of the effective magnetic field  $H_N$  of the polarized nuclei. The nuclear field  $H_N$  is due to the dynamic polarization of the nuclei by the oriented electrons. The general expression for  $H_N$  in the approximation linear in the average electron spin  $S$  takes the form

$$H_{N\alpha} = \sum_{\beta} a_{\alpha\beta} S_{\beta}, \quad (1)$$

where  $a_{\alpha\beta}$  is a tensor whose components depend on the external magnetic field  $H$  and on its direction relative to the crystal axes. We shall assume henceforth that the coordinate axes are directed along the fourfold axes of the crystal. The symmetry properties of this tensor were considered in Ref. 10. It follows from these symmetry properties, in particular, that if the direction of the magnetic field coincides with one of the symmetry axes of the crystal, then expression (1) becomes substantially simpler.

If the field  $H$  is directed along the  $[100]$  axis, only the diagonal components of the tensor  $\hat{a}$  differ from zero:  $a_{zz} = a_{yy}, a_{xx}$ . Then

$$H_N = aS + b(\text{Sh})h, \quad (2)$$

where  $h$  is a unit vector along  $H$ ,  $a = a_{zz}$ ,  $b = a_{xx} - a_{zz}$ . If  $H$  is directed along the  $[111]$  axis, then  $a_{xx} = a_{yy} = a_{zz}$ , and all the off-diagonal components are equal. In this case Eq. (2) remains valid, but now  $a = a_x - a_{xy}$ ,  $b = 3a_{xy}$ .

Finally, if  $H$  is parallel to the  $[110]$  axis, then the nonzero components are  $a_{xx} = a_{yy}$ ,  $a_{zz}$ ,  $a_{xy} = a_{yx}$ . In this case

$$H_N = aS + b(\text{Sh})h + \gamma \hat{c}S, \quad (3)$$

where  $a = a_{zz}$ ,  $b = 2(a_{xx} - a_{zz})$ ,  $\gamma = a_{xy} - a_{xx} + a_{zz}$ ;  $\hat{c}$  is a tensor that has two nonzero components:  $c_{xy} = c_{yx} = 1$ .

Equations (1)–(3) are of general character. Without allowance for the quadrupole interactions, the use of the idea of nuclear spin temperature leads to an expression for the nuclear field in the form<sup>11</sup>  $H_N = b(S \cdot h)h$ . The additional terms are connected with the crystal anisotropy. For the case when this anisotropy is due to the quadrupole interactions in the  $\text{Ga}_x\text{Al}_{1-y}\text{As}$  solid solution, the components of the tensor  $a_{\alpha\beta}$  were obtained in Ref. 10 on the basis of the following model. It was assumed that the nuclear field acting on the electron spins consists of two parts:

1. The field produced by the oriented arsenic nuclei

of four types, for each of which the quadrupole-interaction axis is one of the four threefold axes  $\{111\}$ . Each type corresponds to replacement of one of the four nearest gallium atoms by an aluminum atom. These arsenic nuclei will be called the perturbed nuclei.

2. The field produced by the other nuclei, for which the quadrupole splitting is insignificant (unperturbed nuclei).

It follows from the calculations in Ref. 10 that the main contribution to the nuclear field of the perturbed nuclei is made by states with spin projections  $\pm 3/2$  on the corresponding quadrupole-interaction axes (the spin of the arsenic nucleus is  $3/2$ ). The perturbed nuclei in these states produce nuclear fields directed along the corresponding threefold axes. The polarization of the unperturbed nuclei can be characterized by a nuclear spin temperature, and consequently their average spin and the nuclear field produced by them are directed along the external magnetic field.

We begin the analysis of the nonstationary phenomena from the following equation, which describes the time behavior of the nuclear field:

$$\frac{d\mathcal{H}_N}{dt} = -\frac{1}{T_1}(\mathcal{H}_N - H_N), \quad (4)$$

where  $\mathcal{H}_N$  is the instantaneous value of the nuclear field. Equation (4) corresponds to the simplest assumptions concerning the character of the nuclear spin relaxation: it is assumed that both the perturbed and the unperturbed nuclei have the same longitudinal-relaxation time  $T_1$ . This makes it possible to write down a single equation that describes the evolution of the combined nuclear field. (In the general case, when several different relaxation times that characterize the time dependence of the state of the nuclear system, the evolution of the nuclear field is described by a much more complicated system of equations.) In addition, the nuclear polarization is assumed to be small. Otherwise the expression for the nuclear field would have a much more complicated form, and the time  $T_1$  could itself depend on the magnitude and direction of the nuclear field.

We emphasize that the vector equation (4) has a most unusual form. It contains no terms corresponding to precession of the nuclear magnetization around the magnetic field. The reason is that the spins of the perturbed nuclei are always directed along the quadrupole-interaction axes, while the spins of the unperturbed nuclei are directed along the external magnetic field. If only unperturbed nuclei were present, then only the nuclear spin temperature could vary with the time  $T_1$ , and not the direction of the nuclear field (which is always parallel in this case to the external field). On the other hand, for any type of perturbed nuclei, deviation of the spin from the direction of the corresponding quadrupole-interaction axis is also impossible. However, according to Eq. (4) the summary field  $\mathcal{H}_N$  varies not only in magnitude but also in direction with change of the characteristic time  $T_1$ . The possibility of slow rotations of the nuclear field, which

follows from (4), is connected not with the rotations of the individual nuclear spins, but with the change of the polarization of nuclei of various types, whose spin directions are always fixed.

The nuclear field  $\mathcal{H}_N$  together with the external field  $H$  exerts an influence on the magnitude and direction of the electron spin orientation. The electron spin aligns itself practically immediately with the instantaneous value of the nuclear field  $\mathcal{H}_N$ . To determine  $S$  in terms of  $\mathcal{H}_N$  we have an equation that describes the Hanle effect in the combined field  $H + \mathcal{H}_N$ :

$$\frac{S - S_0}{\tau} = \frac{\mu_0 g}{\hbar} (H + \mathcal{H}_N) \times S, \quad (5)$$

where  $\mu_0$  is the Bohr magneton,  $g$  is the  $g$ -factor of the electron,  $S_0$  is the average electron spin produced by the light in the absence of a magnetic field, and  $\tau$  is the time that determines the width of the Hanle curve in the absence of a nuclear field.

The system (4), (5) can be used to study the stability of the stationary states of an optically oriented electron-nuclear system.

We shall use henceforth dimensionless variables and choose the units of the magnetic field to be  $\hbar/\mu_0 g \tau$  (the half-width of the Hanle curve in the absence of a nuclear field), the unit of the average spin  $S$  is taken to be  $S_0$ , and the unit of the time is taken to be the time of the longitudinal nuclear relaxation  $T_1$ . Then Eqs. (1)–(3) remain in force provided the constants  $a_{\alpha\beta}$ ,  $a$ ,  $b$ , and  $\gamma$  are suitably redefined; Eqs. (4) and (5) take the form

$$d\mathcal{H}_N/dt = -(\mathcal{H}_N - H_N), \quad s - k = [H + \mathcal{H}_N, s], \quad (6)$$

where  $k$  is a unit vector in the direction of  $S_0$ , and  $s$  stands for  $S$  measured in units of  $S_0$ .

### 3. INSTABILITIES IN A TRANSVERSE MAGNETIC FIELD

We consider the case when the external magnetic field is perpendicular to the excitation direction (the Hanle effect). We assume for simplicity that the vector  $S_0$  is parallel to the [001] axis of the crystal, and that the magnetic field lies in the (001) plane. This geometry was realized in the experiments of Refs. 2, 3, and 5. As follows from our earlier results,<sup>10</sup> under stationary conditions the projection  $s_z$  of the average spin of the electron on the direction of the excitation is connected with the magnitude of the external magnetic field  $H$  by the equation

$$H^2 = \frac{1 - s_z}{s_z} \frac{(1 + \varepsilon s_z - \alpha s_z^2)^2}{1 + 2\delta s_z + \gamma^2 s_z^2}, \quad (7)$$

where the parameters  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\varepsilon$  are connected in the following manner with the coefficients of the tensor  $a_{ik}$ :

$$\begin{aligned} \alpha &= \alpha_{xy}\alpha_{yz} - \alpha_{xz}\alpha_{yy}, & \varepsilon &= \alpha_{xy} - \alpha_{yz}, \\ \gamma^2 &= (\hbar_y\alpha_{xy} - \hbar_x\alpha_{yy})^2 + (\hbar_y\alpha_{xz} - \hbar_x\alpha_{yz})^2, & & \\ \delta &= \hbar_y(\hbar_y\alpha_{xy} - \hbar_x\alpha_{yy}) + \hbar_x(\hbar_y\alpha_{xz} - \hbar_x\alpha_{yz}), & & \\ a_{ik} &= a_{ik} - a_{zz}\delta_{ik}. & & \end{aligned} \quad (8)$$

The character of the stationary Hanle curve was investigated earlier.<sup>10</sup> If  $h$  is directed along the [100]

axis, then  $\alpha = \varepsilon = \gamma = \delta = 0$  and Eq. (7) leads to the usual form of the Hanle curve  $s_z = (1 + H^2)^{-1}$ . On the other hand if the field is directed along the [110] axis, then  $\varepsilon = \delta = 0$ , but  $\alpha$  and  $\gamma$  are generally speaking not equal to zero and the Hanle curve has a complicated character. At  $\alpha > 1$  or at sufficiently large negative value of  $\alpha$ , a hysteresis region exists on the Hanle curve. The character of the hysteresis is different in these two cases (see Fig. 1). There are three stationary states in the hysteresis region. We investigate now the stability of these states.

We put

$$\mathcal{H}_N = \mathcal{H}_N^{(0)} + \mathcal{H}_N'(t), \quad s = s^{(0)} + s'(t),$$

where  $\mathcal{H}_N^{(0)} = H_N(s^{(0)})$  is the stationary value of the nuclear field,  $s^{(0)}$  is the stationary value of the electron spin, and the perturbations  $\mathcal{H}_N'(t)$  and  $s'(t)$  are proportional to  $e^{\lambda t}$ . Then Eqs. (6) with allowance for (1) and (2) lead to a system of homogeneous linear equations for the three projections of  $\mathcal{H}_N'$ :

$$\lambda \mathcal{H}_N' = -\hat{B} \mathcal{H}_N'.$$

Equating the determinant of this system to zero, we obtain, as usual, a cubic equation for  $\lambda$ . It is important that one of the roots of this equation is known ( $\lambda_3 = -1$ ). In fact, the perturbation  $\mathcal{H}_N'$ , which is directed along the stationary value of the spin  $s^{(0)}$  does not influence the average spin, and therefore  $s' = 0$  for this perturbation, and  $H_N$  remains unchanged:  $H_N = H_N(s^{(0)})$ . For this perturbation we then have  $d\mathcal{H}_N'/dt = -\mathcal{H}_N'$ , i.e.,  $\lambda = -1$ . For the determination of the remaining two roots we obtain the quadratic equation  $\lambda^2 + 2p\lambda + q = 0$ , where  $q = \det \hat{B}$ ,  $2p = \text{Sp} \hat{B} - 1$ . We have

$$\lambda_1 = -p + (p^2 - q)^{1/2}, \quad \lambda_2 = -p - (p^2 - q)^{1/2}.$$

It is seen that if  $q < 0$  then  $\lambda_1 > 0$  for any  $p$ , and consequently the stationary state is unstable.

It is easy to show that  $q$  vanishes only at the points  $C$  (Fig. 1), where  $dH/ds_z^{(0)} = 0$ . In fact, at  $q = 0$  one of the roots,  $\lambda_1$  or  $\lambda_2$ , vanishes. Consequently there should exist an (infinitesimally small)  $s'$  such that  $s^{(0)}$  and  $s^{(0)} + s'$  correspond to a stationary state at one and the same magnetic field  $H$ . Since  $s_x^{(0)}$  and  $s_y^{(0)}$  are single-valued functions of  $H$  and  $s_z^{(0)}$ , this is possible only under the condition  $dH/ds_z^{(0)} = 0$ .

We show now that in the case  $\alpha > 1$  the branch 3 is always unstable. For this it suffices to verify that on this branch we have  $q < 0$  at  $H = 0$ .<sup>1)</sup> Using Eqs. (1)

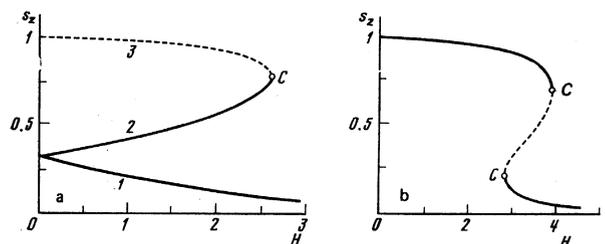


FIG. 1. Hysteresis on the Hanle curve. Pumping along the [001] axis (the  $z$  axis), the field  $H$  is parallel to the [110] axis. a)  $\alpha > 1$ , b)  $\alpha < 0$  (for the hysteresis conditions see Ref. 10). The dashed curves show the branches known to be unstable (in accord with the criterion  $q < 0$ ).

and (6) we can obtain for  $q$  at  $H=0$ ,  $s_x^{(0)}=1$ ,  $s_y^{(0)}=s_z^{(0)}=0$  (branch 3) the general expression

$$q = (1 + \varepsilon - \alpha) / (a_{zz}^2 + 1)$$

for  $H$  directed along  $[110]$  we have  $\varepsilon = 0$  and

$$q = (1 - \alpha) / (1 + a_{zz}^2). \quad (9)$$

We see therefore that at  $\alpha > 1$  we have  $q < 0$ . Consequently  $q < 0$  on the entire branch 3 of Fig. 1a, and this entire branch is unstable. In the case  $\alpha < 0$  it can be proved similarly that branch 2 of Fig. 1b is unstable. In fact, from (9) it follows that at  $\alpha < 0$  the parameter  $q > 0$  at the point  $H=0$ . Consequently  $q > 0$  on the entire branch 3 of Fig. 1b. At the point C, the parameter  $q$  reverses sign, so that on branch 2 of Fig. 1b we have  $q < 0$ , meaning that this entire branch is unstable at  $\alpha < 0$ .

We note that from the arguments presented above it follows that  $q > 0$  on branches 1 and 2 in the case of Fig. 1a and on branches 1 and 3 in the case of Fig. 1b. In addition,  $q < 0$  on the entire Hanle curve in the absence of hysteresis. Consequently, to check on the stability in these cases it suffices to determine the sign of the parameter  $p$ . (The stationary state is stable at  $p > 0$  and unstable at  $p < 0$ ).

We consider first the simplest case when the field  $H$  is directed along the  $[100]$  axis. The nuclear field is then determined by Eq. (2), the Hanle curve has the usual form, and the nuclear field does not influence the stationary value of the electron orientation.<sup>2)</sup> Nonetheless, as we shall presently show, it can lead under certain conditions to instability of the stationary state.

Calculation leads for this case to the following value of the parameter  $p$ :

$$p = \frac{(1 + H^2)^2 - ab/2}{(1 + H^2)^2 + a^2}. \quad (10)$$

We see that instability takes place at  $ab > 2(1 + H^2)^2$ . Since the stationary state is unique in this case, its instability means that undamped oscillations should occur in the bound system of electron and nuclear spins. These oscillations should exist in a zero at  $ab > 2$  and vanish at a sufficiently large value of the magnetic field  $H^2 > (ab/2)^{1/2} - 1$ . The period of these oscillations is determined by the time of the longitudinal nuclear relaxation  $T_1$ . Figure 2 shows the time dependences of  $s_z$  obtained by computer solution of Eqs. (6) and (2).

The physical cause of the instability can be described in the following manner. In the stationary state the electron spin is directed perpendicular to the external field  $H$ , so that the nuclear field is parallel to the electron spin [see (2)] and therefore acts on this spin. However, the fluctuation deviation of the nuclear field from this direction leads to the appearance of an  $s$  component along  $H$ . This in turn leads to a buildup of nuclear spins along  $H$  and by the same token to a still larger deviation of the nuclear field from the initial direction.

If the magnetic field  $H$  is not directed along the  $[100]$  axis, then the stationary Hanle curve does not have a simple Lorentz shape. The expression for the para-

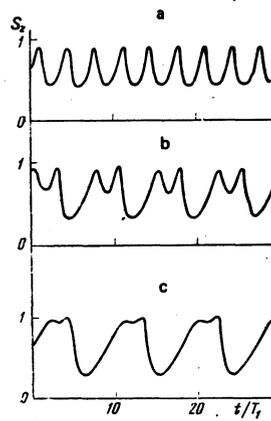


FIG. 2. Oscillations of the average projection of the electron spin  $s_z$  (which determines the polarization of the recombination radiation) under instability conditions in a transverse magnetic field  $a=1$ ,  $b=5$ . The values of the magnetic field  $H$  (in units of the half-width of the Hanle curve) are: a— $H=0$ , b— $H=0.5$ , c— $H=0.75$ .

meter  $p$  is cumbersome in this case and will not be presented here. A simple expression for  $p$  can be obtained for an arbitrary direction of the magnetic field as  $H \rightarrow 0$ :

$$p = \frac{2 - a_{zz}(\alpha_{zz} + \alpha_{yy}) + \varepsilon}{2(1 + a_{zz}^2)}. \quad (11)$$

It is seen that the instability condition  $p < 0$  contains in the general case a parameter  $\varepsilon$  that reverses sign when the direction of the magnetic field relative to the axes  $[110]$  or  $[100]$  is reflected, or when the sign of the circular polarization of the exciting light is reversed.<sup>10</sup> Therefore the conditions for the onsets of instability and of oscillations are generally speaking different when the vector  $H$  lies on opposite sides of the twofold and fourfold axes.

The influence of the electron fields leads to an additional distortion of the stationary Hanle curve, and in particular to the appearance on it of an additional maximum at  $H \sim (H_N H_e)^{1/2}$ , where  $H_N$  and  $H_e$  are constants that characterize respectively the magnitudes of the electron and nuclear fields.<sup>10,12</sup> At this maximum, the nuclear field cancels the external field  $H$ , so that the total transverse field acting on the electron spin is close to zero. One should expect a negative value of expression (11) to serve as the criterion precisely at this point.

The undamped slow (with period of the order of several seconds) oscillations of the degree of polarization of the luminescence under optical orientation in  $\text{Ga}_{1-x}\text{Al}_x\text{As}$  solid solutions in a perpendicular magnetic field were observed by Novikov and Fleisher.<sup>3</sup> They have also found that the region of existence of oscillations depends substantially on the sign of the circular polarization of the exciting light. These results agree qualitatively with the developed simple theory based on Eqs. (6) and on symmetry considerations. To obtain quantitative results, however, and in particular to determine the interval of directions and magnitudes of the field  $H$  in which oscillations exist, these arguments are insufficient, and further information is needed on the

values of the parameters and on their dependence on  $H$ . In Refs. 3 and 5 the oscillations were observed for a magnetic field directed in the vicinity of the  $[110]$  axis and in the intermediate vicinity of the hysteresis region.

#### 4. INSTABILITY IN A LONGITUDINAL MAGNETIC FIELD

Dzhioev, Zakharchenya, and Fleisher<sup>13</sup> observed a dip on the plot of the degree of polarization of the luminescence as a function of the magnetic field parallel to the excitation direction. The dip was observed in the region of weak fields (of the order of 1 Oe) and was strongly asymmetrical in shape (Fig. 3). The exciting beam and the magnetic field were directed along the  $[111]$  axis of the crystal in these experiments. Under these conditions it might seem that the magnetic field should have no influence on the electron spin. The existence of the dip was attributed in Ref. 13 to the influence of the nuclear field on the rate of the spin relaxation of the electrons. The fact that the dip is shifted away from the point  $H=0$  was attributed to the influence of the electric field on the dynamic polarization of the nuclei. According to this explanation, the minimum on Fig. 3 corresponds to cancellation of the electric field acting on the nuclear spins by the external magnetic field, so that the total magnetic field at the nuclei is equal to zero, and no dynamic polarization of the nuclei takes place.

In this section we show that the dip can be due also to another cause, namely instability of the stationary state in which the electron spin is parallel to the external magnetic field. It is not necessary in this case to assume that the rate of the electron spin relaxation depends on the magnetic field in the weak-field region.

In weak fields the behavior of the nuclear spins can be described with the aid of the nuclear spin temperature  $\Theta$ . For the nuclear field  $H_N$  (which is proportional to the average value of the summary nuclear spin) we then have  $H_N \sim H_N/\Theta$ . With the electron field taken into account,  $H$  should be taken to mean the sum of the external and electron fields, but since the electron field is always directed along the average electron spin  $s$ , and consequently does not affect  $s$ , we can assume that  $H$  is simply the external field.

However, in the considered case of a solid solution, description with the aid of spin temperature is not applicable to all the spin levels of the nuclei. For per-

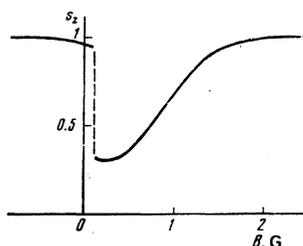


FIG. 3. Dip on the plot of the degree of polarization of the recombination radiation against the longitudinal magnetic field in accord with the experimental data of Ref. 13.

turbed nuclei of arsenic on the levels  $\pm 3/2$ , there is no dipole-dipole interaction with the spins of the surrounding nuclei, since the transitions  $+3/2 \rightleftharpoons 3/2$  are forbidden.<sup>3)</sup> Therefore the perturbed nuclei at the levels  $\pm 3/2$  can be regarded as independent. Then the nuclear field due to them is proportional under stationary conditions to the electron spin  $s$ , and we can write for the nuclear field the expression

$$H_N = as + BH/\Theta. \quad (12)$$

We note that although under stationary conditions the field of the perturbed nuclei is directed along  $s$  and therefore does not affect  $s$ , in the nonstationary regime the direction of this field and the direction of the spin  $s$  may not coincide, a fact that must be taken into account in the investigation of the stability.

The time dependences of the electron and nuclear spins is determined as before by Eqs. (6), where  $H_N$  takes the form (12). The condition for stability of the stationary state, under which the electron spin is parallel to the external field ( $s_x^{(0)} = 0, s_y^{(0)} = 0, s_z^{(0)} = 1, z$  is the direction of the field  $H$ ), takes the form  $p > 0$  (see Sec. 2). The calculations yield

$$(1+c^2)p = 1+c(c-a), \quad c = \mathcal{H}_N^{(0)} + H = a + BH/\Theta^{(0)} + H. \quad (13)$$

Following Ref. 11, we assume that

$$\frac{B}{\Theta} = b \frac{Hs}{H^2 + H_L^2}, \quad (14)$$

where  $b$  is a certain constant, and we replace in (14) the field  $H$  by the sum of the external and electron fields:  $H \rightarrow H + H_e s$ . It is seen that instability can set in only if  $H/\Theta^{(0)} < 0$ , i.e., if the sign of the temperature does not correspond to the sign of the projection of the external field on the spin direction. This situation is due to the electron field. Indeed, if the electron field is directed opposite to the external field and exceeds the latter, then the sign of the combined field acting on the nuclear spins does not coincide with the sign of the external field, and the sign of the spin temperature changes correspondingly. It follows therefore that the instability can arise only if the external magnetic field is antiparallel to the electron field and is smaller than the latter in magnitude. This is in qualitative agreement with experiment (see Fig. 3), according to which the observed dip is asymmetrical with respect to the reversal of the sign of  $H$  at a constant sign of the polarization of the exciting light.

We show now that at values of the magnetic field  $H$  such that the state  $S \parallel H$  is unstable there exists another state in which the electron spin and the nuclear field processes slowly around  $H$ . Substituting (12) in Eqs. (6) we obtain the following system of equations:

$$\begin{aligned} \frac{dA_x}{dt} &= -A_x + H + \left( a + \frac{BH}{\Theta} \right) s_x, \\ \frac{dA_x}{dt} &= -A_x + as_x, \quad \frac{dA_y}{dt} = -A_y + as_y, \end{aligned} \quad (15)$$

where  $A$  denotes the total magnetic field acting on the electron spin:  $A = H + \mathcal{H}_N$ . The components of  $s$  are expressed in terms of  $A$  with the aid of the second equation of (6):

$$s_x = \frac{1+A_z^2}{1+A^2}, \quad s_y = \frac{A_y+A_z A_x}{1+A^2}, \quad s_z = \frac{-A_x+A_y A_z}{1+A^2}. \quad (16)$$

Equations (15) and (16) have, in addition to the stationary solution

$$A_x = H + (a + BH/\theta), \quad A_y = A_z = 0; \quad (17)$$

$$s_x = 1, \quad s_y = s_z = 0$$

also a second solution

$$A_x = A_\perp \sin(\Omega t + \varphi), \quad A_y = A_\perp \cos(\Omega t + \varphi). \quad (18)$$

Here  $\varphi$  is an arbitrary phase,  $\Omega = A_z^{-1}$ , and  $A_z$  and  $A_\perp$  are constants defined by the system of equations

$$aA_z = 1 + A_\perp^2 + A_z^2, \quad aA_z^2 = aA_z H + (a + BH/\theta)(1 + A_z^2). \quad (19)$$

The components of the spin  $s$  are connected with  $A$  by Eqs. (16), so that  $s_z$  is constant, while  $s_x$  and  $s_y$  oscillate with frequency  $\Omega$ .

We assume for simplicity that the external magnetic field is so small that its indirect action on the electron spin can be neglected [i.e., we discard the terms  $aA_z H$  in (19) and  $H$  in (13)]. Let, in addition, both the external and the electron fields be much weaker than the local field  $H_L$ . We then obtain for the second solution

$$s_x = \frac{1}{(c(a-c))^{1/2}}, \quad \Omega = \left(\frac{a-c}{c}\right)^{1/2}, \quad c = a + b \frac{H(H+H_L)}{H_L^2}. \quad (20)$$

The obtained solution exists at  $c(a-c) > 1$ , i.e., in the entire region of magnetic fields where the first solution is unstable [ $p < 0$ , see Eq. (13)].

Figure 4 shows the  $s_z(H)$  dependence that follows from (20) at different values of the parameter  $\eta = bH_0^2/aH_L^2$ . At small values of this parameter the dip has a simple shape (curve 1 on Fig. 4a), and at large values the shape of the curve becomes more complicated (Fig. 4b) and it is possible even to have two dips separated by a region of magnetic-field values such that the first state is stable ( $s_z = 1$ ) (Fig. 4c). Figure 4d shows the result of the calculation in the general case when  $H \sim H_0$ ,  $H_L \sim 1$ .

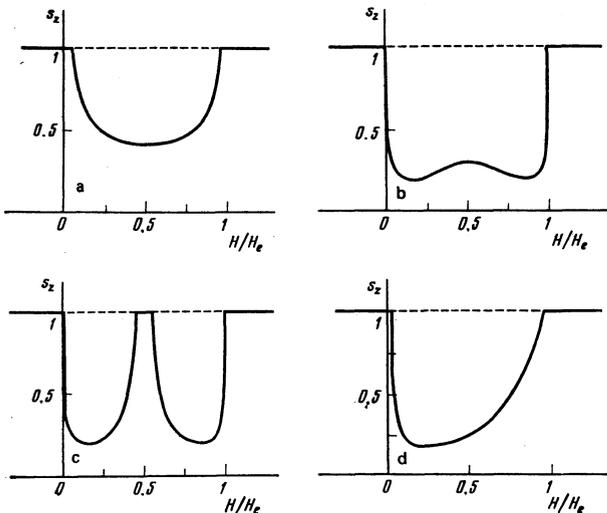


FIG. 4. Dip on the plot of  $s_z$  against the longitudinal field  $H$ . The dashed line shows the stationary state  $s_z = 0$  in the region of its instability. In this region the spin  $s$  and the nuclear field  $\mathcal{H}_N$  precess around the  $z$  axis. Calculation by formula (20) at  $a = 10$  and values of the parameter  $\eta$ : a—0.25, b—3.5, and c—4; d—calculation in accord with the general formulas (14), (16), and (19) at  $a = 30$ ,  $b = 5$ ,  $H_0 = 1$ , and  $H_L = 0.5$ .

It can be assumed that the experimentally observed<sup>13</sup> dip on the  $s_z(H)$  curve is due to the instability described above.

The decrease of  $s_z$  is actually due to the Hanle effect in the nuclear field, which becomes oblique as a result of the instability. Under real conditions there are many complicating circumstances which are not accounted for in the simple model considered here: inhomogeneity of the electron orientation, which leads in particular to inhomogeneity of the electron field, the difference between the relaxation times of nuclei of different sorts, etc.. The phase shifts of the rotation of the nuclear field at different points of the sample can be different. Then the nuclear field produced as a result of the instability can have a random character, and the decrease of the electron orientation can be interpreted as spin relaxation in these random fields. The existence of this relaxation was in fact suggested in Ref. 13.

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<sup>1</sup>By zero field we mean a field much smaller than the half-width of the Hanle curve in the absence of nuclear polarization. This field must nevertheless be much larger than the local field, to be able to regard the parameters  $a_{ik}$  as independent of the field.<sup>10</sup>

<sup>2</sup>We do not take into account here the field produced by the polarized electrons at the nuclei.

<sup>3</sup>In fact, a matrix element of the angular momentum between these states can exist if account is taken of a small admixture of states  $\pm 1/2$ , which is due, for example, to the influence of the aluminum atoms, which leads to a weak non-coaxiality of the quadrupole interaction. (We have proposed the existence of this matrix element in Ref. 10 in order to explain the strong anisotropy of the Hanle curve.) These effects, however, are small and the populations of the levels  $\pm 3/2$  can be described with the aid of the spin temperature only in very weak fields, smaller by one or two orders of magnitude than the local field  $H_N$ . In the case considered here even the electron field alone is sufficient to suppress the interaction of the perturbed nuclei on the levels  $\pm 3/2$  with the surrounding nuclei.

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## $(\text{He}^3)_2$ van der Waals molecular dimers in solutions of the quantum liquids $\text{He}^3$ – $\text{HeII}$

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Bound states of  $\text{He}^3$  impurity quasiparticle pairs, produced as a result of van der Waals attraction on the free surface in thin films, in narrow capillaries, and on vortex filaments in superfluid  $\text{He}^4$  are investigated. With decreasing temperature, the contribution of the bound states to the thermodynamics of the solution becomes decisive. The coefficient of inelastic absorption of first sound due to the bound state in the field of the acoustic wave is calculated. It is predicted that a system of impurity excitations in narrow capillaries or on vortex filaments can have a resonant singularity (at a frequency corresponding to the threshold of the splitting of the bound state) wherein the absorption coefficient for monochromatic sound becomes infinite. The temperature of the gas-liquid phase transition and the superfluid transition temperature are calculated for a Bose system of van der Waals pairs  $(\text{He}^3)_2$  on a surface and in thin  $\text{HeII}$  films. It is shown that the superfluid transition temperature can be of the order of 35 mK.

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It is known that  $\text{He}^3$  atoms dissolved in superfluid  $\text{He}^4$  form, at sufficiently low concentrations, a weakly non-ideal gas with attraction between the quasiparticles.<sup>1–3</sup> The presence of arbitrarily weak attraction between fermions leads at exponentially low temperatures to the formation of Cooper pairs and to a phase transition of the dissolved  $\text{He}^3$  into the superfluid state (see, e.g., Refs. 2, 4, 5). This attraction between the impurity excitations turns out, however, to be too weak to produce Van der Waals bound states of  $\text{He}^3$  particles within the volume of the solution. It appears therefore that no Van der Waals dimer  $(\text{He}^3)_2$  are produced under ordinary conditions. (We note that generally speaking at high pressures, when the effective mass and the radius of the interaction of the  $\text{He}^3$  quasiparticles increase, these bound states might appear.

On the other hand, in a two-dimensional attraction field a discrete  $s$ -level corresponding to a bound state of particles is always present. One can regard as two-dimensional, for example, a system of impurity excitations of  $\text{He}^3$  in a sufficiently thin film of  $\text{HeII}$ , bounded on one or both sides by a solid surface. The  $\text{HeII}$  film can then turn out to be fully macroscopic, i.e., its thickness can greatly exceed atomic dimensions. In fact, let the attraction between the dissolved  $\text{He}^3$  atoms lead in the two-dimensional case to formation of a bound-state  $(\text{He}^3)_2$  with characteristic dimension  $r_0$ . It is then clear that the interaction of the impurity excitations in the  $\text{HeII}$  film can be regarded as two-dimensional if the film thickness  $d$  is much less than the dimer dimension, i.e.,  $d \ll r_0$ . The exponential smallness

of the binding energy  $\Delta$  of the dimer compared with the characteristic kinetic energy  $\hbar^2/ma^2$  ( $m$  is the effective mass of the  $\text{He}^3$  quasiparticle and  $a$  is the atomic dimension) means that  $r_0 \gg a$ , i.e., there exists a region of macroscopic thickness of the film  $a \ll d \ll r_0$ . Since we are confining ourselves only to pair interaction of particles, a procedure valid only for sufficiently low concentrations, such that  $r_0 \ll l$  ( $l$  is the average distance between the dissolved  $\text{He}^3$  atoms), we are dealing in fact with a rarefied monolayer of impurity quasiparticles in a macroscopic film of superfluid  $\text{He}^4$ .

It should also be noted that under the influence of the Van der Waals forces it is  $\text{He}^4$  which crystallizes predominantly on the solid surface that bounds the solution film, owing to the difference between the molar volumes of the helium isotopes. According to the phase diagram of the solid solutions at temperatures  $T \leq 0.318$  K, the  $\text{He}^3$  atoms are practically not dissolved in crystalline  $\text{He}^4$  (see, e.g., Ref. 6). This makes it possible to explain qualitatively why the dissolved  $\text{He}^3$  atoms are not localized on a solid wall, and they can be regarded as a gas of excitations with two-dimensional interaction between them (in this sense, the interaction of the impurity quasiparticles with the solid wall remains three-dimensional).

A model-based calculation of the dependence of the concentration of the dissolved  $\text{He}^3$  atoms on the distance to the solid surface, in a wide temperature interval, was carried out by Peshkov.<sup>7</sup>

Surface impurity  $\text{He}^3$  states on the free surface of a