Spin resonance of nonlinear optical susceptibility in *n*-InSb

M. S. Bresler and O. B. Gusev

A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences

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Spin resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ responsible for a frequency shift of the form $\omega_3 = 2\omega_1 - \omega_2$ is investigated in *n*-InSb in a wide range of conduction-electron concentrations. The experiments were performed with a *Q*-switched CO_2 laser generating simultaneously at two frequencies, $\omega_1 \approx 944$ cm^{-1} and $\omega_2 \approx 1044$ cm^{-1}. The dependence of the resonance position, of the line shape, and of the resonance amplitude on the conduction-electron concentration is well described by the quantum theory of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ based on a solution of the equation for the density matrix with account taken of the motional narrowing of the resonance line. The spin and momentum relaxation times of the conduction electrons are determined.

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INTRODUCTION

The nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$, responsible for frequency mixing of the type $\omega_3 = 2\omega_1 - \omega_2$ is resonantly amplified if the energies ω_1 and ω_2 of the pumping-radiation quanta or the difference of these energies $\omega_2 - \omega_1$ coincide with the energy of one of the three intermediate states through which this four-photon coherent process goes through.

In a narrow-band semiconductor, this characteristic energy may be the energy of the orbital motion of the electron in a magnetic field (Landau quantization energy) or the energy of the Zeeman splitting connected with the spin. Accordingly, the nonlinear optical susceptibility $\chi^{(3)}$ experiences resonance at the cyclotron frequency of the conduction electrons ω_c [henceforth called for brevity cyclotron resonance (CR); cyclotron resonance at double the cyclotron frequency $2\omega_c$ is also possible] or at the spin frequency ω_s of the conduction electron [henceforth spin resonance (SR)].

The CR of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ in the narrow-band semiconductor *n*-InSb was investigated by us previously.[1]

In the present paper we report the results of the investigations of SR of the nonlinear optical susceptibility in *n*-InSb.

The resonant susceptibility of the frequency mixing of the type $\omega_3 = 2\omega_1 - \omega_2$ is closely connected with the susceptibility responsible for the coherent Raman scattering,[2] in this case for Raman scattering with spin flip, which has been investigated in very great detail.[3-5]

Spin resonance of nonlinear optical susceptibility was first observed $\Delta\omega = \omega_2 - \omega_1$,[6-8] in the process of generation of the difference frequency, and also in a frequency-mixing process of the type $\omega_3 = 2\omega_1 - \omega_2$, both with the aid of a CO laser[9-11] and with the aid of a CO_2 laser.[12]

The difference between the SR of the nonlinear susceptibility and the CR considered in Ref. 1 is that the former is observed in substantially stronger magnetic fields than CR. First, the spin splitting of the electron levels is usually weaker than the orbital splitting (by approximately a factor of three in the *n*-InSb); second, it is more convenient to observe the CR at the doubled frequency rather at the fundamental one, so that the magnetic fields now differ by a factor of six. Therefore the quasiclassical description ($\hbar\omega_c \ll \xi_F$), which is suitable for CR and makes it possible, for example, to use the method of the equations of motion (see Ref. 1), is of little use for the description of SR. In the case of SR it is necessary to take into account the quantization of the electron spectrum and to solve an equation for the density matrix.

THEORY

In a narrow-band semiconductor whose energy spectrum is described by the Kane model, allowance for the spin-orbit interaction in the one-band Hamiltonian for the conduction band produces an energy term

$$\frac{c}{\varepsilon_g}\mu\left[\mathbf{E}\times\left(\mathbf{p}-\frac{e}{c}\mathbf{A}\right)\right]\frac{\Delta+2\varepsilon_g}{\Delta+\varepsilon_g}, \tag{1}$$

which is responsible for the combined resonance.[13]

Here $\mu = 1/2 g\mu_B\sigma$ is the magnetic moment of the conduction electron, g is the spectroscopic-splitting factor of the band electron, Δ is the energy of the spin-orbit splitting, **E** is the electric field, and **A** is a vector potential that can include not only the vector potential of the field **E**, but also the vector potential of the constant magnetic field **H**. Thus, expression (1) contains a nonlinear term, and it is this term which causes the spin-dependent nonlinearity for the electron in the Kane band.

We specify the vector potential of the electromagnetic wave in the form

$$\mathbf{A} = \sum \mathbf{A}_j e^{i(\mathbf{q}_j\mathbf{r}-\omega_j t)} + \text{c.c.} \tag{2}$$

We shall not bother with a suitable normalization of the vector potential, since we shall be interested hereafter only in the relative (to the nonresonant) magnitude of the resonant optical susceptibility. We shall use also the dipole approximation, i.e., $\mathbf{q}_j \to 0$.

A more accurate expression for the nonlinear part of the spin-orbit contribution to the energy is

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$$\mathscr{H}_{s0}=ia\boldsymbol{\sigma}[\mathbf{A}_1\times\mathbf{A}_2^*], \tag{3}$$

where

$$a=\left(\frac{e}{c}\right)^2\frac{2P^2}{3m_0\varepsilon_g}\left[\frac{\varepsilon_g^2}{\varepsilon_g^2-(\hbar\bar{\omega})^2}-\frac{\varepsilon_g^2}{(\varepsilon_g+\Delta)^2-(\hbar\bar{\omega})^2}\right]\frac{\hbar\bar{\omega}}{\varepsilon_g}, \tag{4}$$

P is the matrix element of the Kane theory, m_0 is the mass of the free electron , and $\bar{\omega}=1/2(\omega_1+\omega_2)$.

The operator $\mathscr{H}_{s0}$ is obtained after summing the second-order matrix element responsible for the Raman scattering with spin flip over the interband transitions; it was used to describe the generation of the difference frequency[7] $\omega_2-\omega_1$ and the frequency mixing.[11] In expression (3), $\mathscr{H}_{s0}$ is already expressed in a form that accounts for the frequency mixing of interest to us, i.e., it depends on the frequency like $\omega_2-\omega_1$. Because of the introduction of this two-photon interaction, our four-photon process of frequency mixing proceeds only via one intermediate state, which in fact turns out to be resonant at $\omega_2-\omega_1=\omega_s$.

We now calculate the nonlinear current at the mixing frequency $\omega_3=2\omega_1-\omega_2$. The nonlinear optical susceptibility $\chi^{(3)}(-\omega_3,\omega_1,\omega_1,-\omega_2)$ is defined by the relation

$$j_\alpha(\omega_3)=i\omega_3\chi^{(3)}_{\alpha\beta\gamma\delta}(-\omega_3,\omega_1,\omega_1,-\omega_2)E_\beta(\omega_1)E_\gamma(\omega_1)E_\delta(-\omega_2). \tag{5}$$

The equation for the conduction-electron density matrix is

$$i\hbar\dot{\rho}=[\mathscr{H},\rho]+i\hbar\left.\frac{\partial\rho}{\partial t}\right|_{coll}, \tag{6}$$

where the density matrix is considered in the Landau-function representation, $\mathscr{H}=\mathscr{H}_0+\mathscr{H}_{s0}$, $\mathscr{H}_0$ is the kinetic-energy operator in a static magnetic field with allowance for the nonparabolicity, and the solution is sought in the form $\rho=\rho_0+\rho$, where ρ_0 is the equilibrium density matrix (we consider hereafter only a Fermi-degenerate electron system).

The solution for the off-diagonal component of the density matrix is

$$\rho_{\alpha\beta}'=\frac{\hbar^{-1}(\mathscr{H}_{s0})_{\alpha\beta}(\rho_{0\alpha}-\rho_{0\beta})}{\Delta\omega-\omega_{\beta\alpha}-i\tau_s^{-1}}, \tag{7}$$

where

$$\Delta\omega=\omega_2-\omega_1\ (>0),\qquad \omega_{\beta\alpha}=\omega_s(k_z)=[\varepsilon_\beta(k_z)-\varepsilon_\alpha(k_z)]/\hbar,$$

τ_s is the electron spin relaxation time.

It is assumed that the density matrix has been summed over the continuous quantum number k_y, while α and β number states with different Landau quantum numbers and with different spin directions. We shall actually consider only the two-level situation close to the quantum limit, i.e., a Landau quantum number $N=0$, and α and β states with spins ↑ and ↓ (the g-factor of n-InSb is negative and the ground state is α). We have furthermore

$$\mathbf{j}=\operatorname{Sp}\widetilde{\rho\mathbf{j}}=\sum_{\alpha\beta}\rho_{\alpha\beta}'j_{\beta\alpha}. \tag{8}$$

We define the current operator with the aid of the expression[14]

$$\delta\mathscr{H}_{s0}=-\frac{1}{c}\hat{\mathbf{j}}\delta\mathbf{A}, \tag{9}$$

and

$$\hat{\mathbf{j}}(\omega_s)=-\frac{c}{\omega_1}a[\boldsymbol{\sigma}\times\mathbf{E}_1]. \tag{10}$$

From this we get the nonlinear current connected with the spin-orbit interaction

$$\mathbf{j}(\omega_3)=\sum_{\alpha\beta}\frac{ia^2c^4\omega_1^{-2}\omega_2^{-1}\boldsymbol{\sigma}_{\alpha\beta}[\mathbf{E}_1\times\mathbf{E}_2^*][\boldsymbol{\sigma}_{\beta\alpha}\times\mathbf{E}_1](\rho_{0\alpha}-\rho_{0\beta})}{\Delta\omega-\omega_{\beta\alpha}-i\tau_s^{-1}}. \tag{11}$$

The summation is over the spin states ↑ and ↓; in addition, integration with respect to the continuous quantum numbers k_z and k_y is implied.

Expression (11) contains explicitly only the term corresponding to the resonance at the frequency $\Delta\omega=\omega_{\beta\alpha}$; the antiresonant term has been left out, since it contributes only to the background and is of no interest to us [we shall compare the resonant part of the susceptibility $\chi^{(3)}$ with the nonresonant part in the absence of a magnetic field [(see Ref. 1)].

In the case of weak nonparabolicity or slight occupation of the band, we can put $\omega_{\beta\alpha}\approx\omega_{s0}$, where ω_{s0} is the spin frequency for the bottom of the conduction band, and the resonance line has a Lorentz shape with half-width $2/\tau_s$. In the case of high occupation of the conduction band, the $\omega_s(k_z)$ dependence must be taken into account and the resonance line will be subjected to a broadening analogous to the inhomogeneous broadening known in the theory of magnetic resonance.

The polarization of the radiation at the mixing frequency ω_3 is determined by the vector products in expression (11) for the current, and can be shown to coincide with the polarization of $\mathbf{E}_2$. To observe SR it is thus necessary that the fields with frequencies ω_1 and ω_2 be noncollinear. It will be assumed henceforth in the present paper that $\mathbf{E}_1\parallel\mathbf{H}$ and $\mathbf{E}_2\perp\mathbf{H}$, and then $\mathbf{E}_3\perp\mathbf{H}$.

Starting from expression (11), it is convenient to regard as a characteristic of the resonant part of the nonlinear optical susceptibility the function

$$\langle(\Delta\omega-\omega_s(k_z)-i\tau_s^{-1})^{-1}\rangle, \tag{12}$$

where the angle brackets denote the averaging

$$\langle F\rangle=\int F(\rho_{0\alpha}-\rho_{0\beta})dk_z\Big/\int(\rho_{0\alpha}-\rho_{0\beta})dk_z$$

which reduces, in the case when both levels α and β are occupied, to the usual averaging

$$\langle F\rangle=\int F\rho_{0\alpha}dk_z\Big/\int\rho_{0\alpha}dk_z$$

in the quantum limit.

We have so far disregarded, however, the electron momentum relaxation. While this relaxation does not lead to loss of the total spin of the electron system, it does influence strongly the shape of the SR line. As shown by Brueck and Blum[15] and by Brueck, Mooradian and Blum,[5] in the case when the electron momentum relaxation is much faster than the spin relaxation, the momentum relaxation processes intermix the particles in states with different k_z, so that the precession frequencies of the electrons that participate in the resonance are effectively averaged. As a result, the line that is inhomogeneously broadened (because of the $\omega_s(k_z)$ dependence) becomes narrower and again assumes a Lorentz shape when the condition $\tau_p\Delta\omega_S<1$ ($\Delta\omega_s$ is the smearing, over the spin frequencies, for all the electrons that participate in the resonance, and τ_p is the momentum relaxation time) is satisfied. the width of this Lorentz line is much less than $\Delta\omega_s$ and is directly proportional to τ_p—this is called motional narrowing.

To take into account in our case the electron momentum relaxation, we use for the collision integral the expression of Brueck and Blum,[15] which we modify to allow for the case when both levels α and β are occupied:

$$\left.\frac{\partial\rho}{\partial t}\right|_{coll} = -\frac{\rho_{\alpha\beta}'}{\tau_s} - \frac{1}{\tau_p}[\rho_{\alpha\beta}' - (\rho_{0\alpha} - \rho_{0\beta})\bar{\rho}_{\alpha\beta}'(k_z)], \tag{13}$$

where

$$\bar{\rho}_{\alpha\beta}'(k_z) = \int dk_z \rho_{\alpha\beta}(k_z) \Big/ \int (\rho_{0\alpha} - \rho_{0\beta})\, dk_z. \tag{14}$$

The solution of Eq. (6) with (13) and (14) taken into account leads to a resonant function of the type

$$\begin{gathered}\Omega/(1+i\Omega/\tau_p),\\ \Omega = \langle(\Delta\omega - \omega_s(k_z) - i\tau^{-1})^{-1}\rangle, \quad \tau^{-1} = \tau_p^{-1} + \tau_s^{-1}.\end{gathered} \tag{15}$$

As $\tau_p \to \infty$ this expression reduces to (12) above. For a parabolic band $\omega_s = \omega_{s0}$ and does not depend on k_z; the corrections that depend on τ_p in the numerator and denominator of (25) then cancel each other, and the resonant function yields, as expected, a Lorentz line with a half-width determined only by τ_s and independent of τ_p. In the general case the analysis of (15) calls for numerical integration. Expression (15) was used in Refs. 15 and 5 to analyze the shape of the Raman-scatterine line in n-InSb with spin flip.

The function (15), apart from a coefficient, is the resonant part of the nonlinear optical susceptibility $\chi^{(3)}$-$(-\omega_3, \omega_1, \omega_1, -\omega_2)$ and contains both the real and imaginary parts.

In experiment one measures the radiation power at the mixing frequency $\omega_3 = 2\omega_1 - \omega_2$:

$$P(\omega_3) \sim |\mathrm{j}(\omega_3)|^2 \sim |\chi^{(3)}|^2 = \chi_{nr}^2 + 2\chi_r'\chi_{nr} + \chi_r'^2 + \chi_r''^2, \tag{16}$$

where χ_{nr} is the nonresonant (background) part of the nonlinear optical susceptibility, and is pure real (the characteristic frequencies χ_{nr} greatly exceed the spin-resonance frequency); χ_r' and χ_r'' are the real and imaginary parts of the resonant contribution to the susceptibility. The second term of the expression for $P(\omega_3)$ describes the interference between the resonant contribution with the nonresonant background (it depends on the real part of χ_r), and at $\chi_r \ll \chi_{nr}$ it describes completely the resonant behavior of the nonlinear susceptibility (see Ref. 1). Since the real part of the resonant contribution χ_r' has a dispersion properties, we shall refer to a dispersion part of the resonant function. In the case of spin resonance at relatively small concentrations of the conduction electrons we have $\chi_r \gg \chi_{nr}$ and the main contribution to the resonance is made by the quantities $\chi_r'^2$ and $\chi_r''^2$.

The pump power, the mode structure of the beam, and a number of other factors are not constant in different experiments, so that when theory is compared with experiment it is convenient to use the ratio of the power $P(\omega_3, H)$ at the frequency ω_3 in a magnetic field to the power $P(\omega_3, 0)$ without a magnetic field:

$$\frac{P(\omega_3, H)}{P(\omega_3, 0)} = 1 + 2\frac{\chi_r'}{\chi_{nr}} + \frac{\chi_r'^2 + \chi_r''^2}{\chi_{nr}^2}.$$

We estimate now the ratio of the resonant part of the nonlinear susceptibility to the nonresonant part. For this purpose it is convenient to use the quasiclassical expression for the nonresonant and resonant contributions to the nonlinear current. The expression for the nonlinear current in the quasiclassical approximation can be obtained by solving the equations for translational and spin motion of a conduction electron in InSb, which are coupled by a spin-orbit interaction of the type (1):

$$\begin{gathered}\mathrm{j}(\omega_3) = -\frac{en}{m^{*2}\varepsilon_g}\frac{ie^3E_{1z}^2E_{2x}}{\omega_1^2\omega_2} + \frac{en}{m^{*2}\varepsilon_g}\frac{ie^3E_{1z}^2E_{2x}}{\omega_1^2\omega_2}\cdot\\ \cdot\frac{p_z^{(0)^2}}{2m^*\varepsilon_g}\frac{8\omega_{c0}\omega_c'}{(\Delta\omega - i\tau_p^{-1})^2 - \omega_c'^2} + \frac{en}{m_s^2\varepsilon_g}\frac{ie^3E_{1z}^2E_{2x}}{\omega_1^2\omega_2}\cdot\\ \cdot\frac{\hbar\omega_1}{\varepsilon_g}\frac{\omega_s(\omega_1 + \omega_2)}{(\Delta\omega - i\tau_s^{-1})^2 - \omega_s^2},\end{gathered} \tag{17}$$

where n is the concentration of the conduction electrons,

$$\omega_{c0} = -eH/m^*c, \quad \omega_{s0} = g_0\mu_B H/\hbar, \quad m_s = 2m_0/g_0,$$

$$\omega_c' = \omega_{c0}\left(1 - \frac{2p_\perp^{(0)^2}}{m^*\varepsilon_g} - \frac{p_z^{(0)^2}}{m^*\varepsilon_g} + \frac{2\mu_0 H}{\varepsilon_g}\right)$$

$$\omega_s = \omega_{s0}\left(1 - \frac{p_z^{(0)^2}}{m^*\varepsilon_g} + \frac{2\mu_0 H}{\varepsilon_g}\right),$$

$$E_{1z} = E_z(\omega_1), \quad E_{2x} = E_x(-\omega_2).$$

The first term in (17) describes the nonresonant part of the nonlinear optical susceptibility, which differs somewhat from that obtained in Ref. 1, since the square root of the Kane dispersion law was expanded in (17) in a series (case of weak nonparabolicity). The second term describes CR at the fundamental cyclotron frequency (in contrast to that considered in Ref. 1), and will be of no interest hereafter, while the third term describes SR. All the quantities that depend on the electron-momentum components $p_z^{(0)}$ and $p_\perp^{(0)}$ must be averaged with the Fermi distribution function, just as in Ref. 1.

An estimate of the ratio ξ of the resonant contribution to the nonresonant background from expression (17) yields

$$\xi \sim \left(\frac{m^*}{m_s}\right)^2 \frac{\hbar\varpi}{\varepsilon_g}\varpi\tau_s \gg 1.$$

EXPERIMENTAL RESULTS AND DISCUSSION

The experimental set up and the sample preparation are described in Ref. 1. The optical system in the SR experiments differ from the CR case in that the laser beam was into two beams, in one of which the polarization of the pump radiation was rotated through 90° with the aid of a CdS half-wave plate. The beams were then superimposed and focused on the n-InSb sample. This resulted in the experimental geometry ($\mathbf{E}_1 \perp \mathbf{E}_2$, $\mathbf{E}_1 \| \mathbf{H}$, $\mathbf{E}_2 \perp \mathbf{H}$) needed for the observation of SR. In the control experiments we separated in each of the beams the radiation of only one frequency, ω_1 or ω_2, with the aid of narrow-band dispersion filters. However, since the filter transmission was small and an appreciable fraction of the pump power was lost as a result, no filters were used in most measurements, and each beam contained radiation at both frequencies.

The polarization of the outgoing radiation with frequency ω_3 was verified in control experiments with the aid of a grating polarizer and agreed with the expected polarization ($E_3 \perp H$). In experiments in which the pump radiation was not filtered, the polarizer was likewise oriented in such a way that only the $E_3 \perp H$ component was registered.

The measurements of the SR of the nonlinear optical susceptibility $\chi^{(3)}$ were made on n-InSb samples with concentrations from 8×10^{14} to 6.5×10^{16} cm^{-3} and with mobilities from 4.0×10^5 to 6.5×10^4 cm^2/V-sec. Most samples were so oriented that the magnetic field H was directed along the [100] or [111] axis; some of the samples were not oriented. In contrast to CR, in the SR experiments we observed a weak anisotropy of the g-factor at magnetic-field orientations H∥[100] and H∥[111].

Typical plots of the SR signal [of the power $P(\omega_3)$ at the mixing frequency] against the magnetic field for samples with different electron concentrations are shown in Fig. 1a (curves 1, 2, 3). In weakly doped samples one can clearly see a series of narrow incompletely resolved SR lines, due to CO_2-laser emission at different frequencies of the rotational spectrum of the CO_2 molecule. A similar spectrum of the SR of the nonlinear optical susceptibility was observed by us in Ref. 12, where the experimental data were compared with a calculation based on the band spectra of InSb and on the data on rotational spectra of CO_2 molecule, and where all the lines of the observed spectrum were identified. In particular, the central line of the spectrum corresponds to a frequency difference $\omega_2 - \omega_1 = 102.7$ cm^{-1}, and the remaining lines are separated 1.8 cm^{-1} in frequency.

With increasing electron concentration, the resonant series of the lines shifts towards stronger magnetic fields and the resolution of the individual lines becomes progressively worse; finally, in samples with concentration higher than 3.5×10^{16} cm^{-3}, the rotational structure is completely smeared out, and the purely resonant line acquires a partial dispersion character. The resonance amplitude referred to the nonresonant background decreases sharply with concentration, from values of the order of several dozen to values of order of unity. Figure 1b (curves 1′, 2′, 3′) shows the resonance lines calculated with a computer by the method described at the end of this section. It is seen that the calculation is in satisfactory agreement with the observed shape of the resonance line.

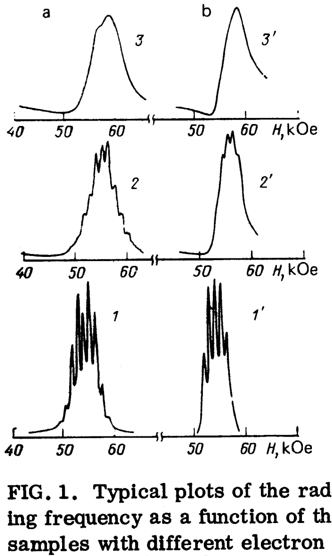

FIG. 1. Typical plots of the radiation power $P(\omega_3)$ at the mixing frequency as a function of the magnetic field, for n-InSb samples with different electron concentration: a) experiment: $1 - n = 7.9\times10^{15}$ cm^{-3}; $2 - n = 2.9\times10^{16}$ cm^{-3}; $3 - n = 4.6\times10^{16}$ cm^{-3}; b) calculation: $1 - n = 8.3\times10^{15}$ cm^{-3} ($\varepsilon_F = 10$ meV); $2 - n = 2.7\times10^{16}$ cm^{-3} ($\varepsilon_F = 21$ meV); $3 - n = 4.3\times10^{16}$ cm^{-3} ($\varepsilon_F = 28$ meV).

The dependence of the position of the SR on the Fermi energy of the electrons is shown in Fig. 2. The Fermi energy was calculated from the electron concentration by a formula that takes into account the nonparabolicity:

$$\varepsilon_F = (\varepsilon_g/2)\{[1 + (2\hbar^2/m^*\varepsilon_g)(3\pi^2 n)^{2/3}]^{1/2} - 1\}.$$

The position of the SR in pure samples was assumed to be the position of the maximum of the central line with maximum intensity out of the series of lines corresponding to the rotational structure. In samples with maximum electron concentration, where the rotational structure was not resolved, the position of the resonance was assumed to be the position of the maximum of the broad resonance line. In the case of intermediate concentrations, where the line did not have a single maximum but the rotational-structure lines were also unresolved (for example Fig. 1a, curve 2), the position of the resonances taken to be the midpoint of the segment drawn at half the height of the resonant peak.

In pure samples, the experimentally observed position of the resonance changes little with the Fermi energy; starting at approximately $\varepsilon_F \approx 10$ meV, the resonance line begins to shift rapidly towards stronger magnetic fields. The light and dark circles in Fig. 2 represent data for samples with orientations [100]∥H and [111]∥H, respectively. These results point to anisotropy ~1.7% of the g-factor at 60 kOe, in accord with the published data.[16] It is known that the anisotropy of the g-factor of the conduction electrons in InSb is due to the interaction of the conduction band with more remote bands.

The singularities of the SR in samples with different electron concentrations can be qualitatively explained with the aid of the energy-level scheme shown in Fig. 3. In the purest samples, the SR takes place in a magnetic field $H_0 = \hbar\Delta\omega/g_0\mu_B \approx 55$ kOe, where $g_0 \approx 47$ is the spectroscopic splitting factor for the bottom of the conduction band. For practically all the samples investigated in the present study, in such strong magnetic fields only

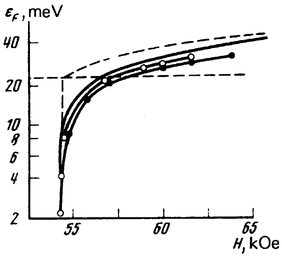

FIG. 2. Position of spin resonance of nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ as a function of the Fermi energy of the condition electrons in n-InSb. Points—experiment: ○ – [100]∥H, ● – [111]∥H. Dashed and solid curves—calculation.

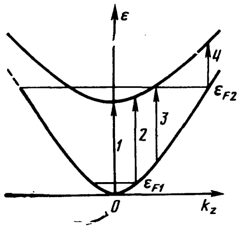

FIG. 3. Transition scheme in spin resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ for different occupation levels of the conduction band of InSb.

the lower Landau subband α (spin ↑)—the quantum limit—was filled, or else both subbands α and β (spins ↑ and ↓) with quantum number $N=0$ were filled. In the presence of spin resonance, transitions from the α band to the β band take place, and because of the nonparabolicity of the energy spectrum the spin frequency ω_s depends on the wave number k_z, i.e., it is different for electrons at different points of the subband. In the quantum limit, when only the band α is filled, the main contribution to the resonance is made by states near the point $k_z=0$ (the transition indicated by the arrow 1 in Fig. 3), since the point $k_z=0$ corresponds to the maximum state density (the square-root divergence in the state density is $k_z=0$ is eliminated on account of the relaxation time τ_s). At low concentrations, only states very close to $k_z=0$ are filled, and for all of them we can put $\omega_s(k_z) \approx \omega_s(0) = \omega_{s0}$. In this case resonance takes place at the frequency ω_{s0}, the line has a Lorentz shape with half-width $2/\tau_s$, and the amplitude is proportional to τ_s.

At higher concentration (Fermi energy ε_{F1} in Fig. 3), one can no longer neglect the $\omega_s(k_z)$ dependence. If we disregard for the time being the motional narrowing of the resonance line, then the qualitative picture of the resonance is the following. The main contribution is made as before by states close to $k_z=0$, so that the maximum of the resonance line remains at the previous value H_0 and the position of the resonance does not change with electron concentration, so long as the Fermi energy remains below the bottom of the upper Landau subband β. States with k_z close to the limiting value k_{zF}, however, produce resonance even at magnetic fields close to H_0, so that the g-factor decreases with k_z because of the nonparabolicity and therefore the resonance line broadens. The $\omega_s(k_z)$ dependence plays in the case of a nonparobolic band the same role as inhomogeneous broadening in the theory of magnetic resonance. The resonance line will have a sharp boundary on the strong-field side at the magnetic field corresponding to $\omega_s(k_{zF})$ (transition 2 in Fig. 3).

Finally, when the electron concentration becomes so large that the upper Landau subband β begins to be filled, states near the point $k_z=0$, i.e., regions with maximum state density become excluded from the region of allowed transition by the Pauli principle. The transitions for a concentration characterized by the Fermi energy ε_{F2} indicated in Fig. 3 can occur only for points of the lower subband, located between the arrows 3 and 4, which mark the limits of the resonance line on the weak and strong fields, respectively. Owing to the square-root energy dependence of the state density, however, the contribution from states near the transition 3 will be larger and the resonance line should have an asymmetric shape with a maximum corresponding to the transition 3. With increasing Fermi energy, the position of the initial point on the Fermi level, for which the transition 3 is allowed, shifts strongly towards higher energies, so that starting with the instant when the Fermi level reaches the upper subband of the point $k_z=0$, the position of the SR as a function of the Fermi energy is strongly shifted towards stronger fields. This agrees qualitatively with the experimentally observed dependence of the SR position on the Fermi energy, but the quantitative description of the experimental data on the basis of such a simple scheme turns out to be unsatisfactory.

The dashed curve in Fig. 2 shows the theoretical dependence of the position of the SR on the Fermi energy. The calculation was performed at the following values of the energy-spectrum parameters: mass at the bottom of the conduction band $m^* = 0.014406 m_0$, $\varepsilon_g = 0.2355$ eV, $g_0 = 47.4$ (these quantities correspond to the band parameters taken from the paper of Pidgeon and Brown,[17] with the effective mass and the g-factor containing a correction for the mass and g factor of the free electron, while the spin-orbit splitting energy is assumed to be $\Delta = 0.92$ eV). The energy $\hbar\Delta\omega$ of the resonant transition was assumed to be 12.785 meV (for a central line with frequency 102.7 cm^{-1}).

The calculations were performed with a computer, using first the given value of the electron Fermi energy (in the absence of a magnetic field) to determine the concentration and the Fermi energy in a magnetic field (the initial magnetic field was specified to be $H_0 = \hbar\Delta\omega / g_0 \mu_B = 54.3$ kOe).

The Fermi energy in a magnetic field was obtained from the equation for a system of electrons in which only the two Landau subbands α and β are filled, and the degeneracy is assumed to be complete (i.e., $T=0$):

$$n = eH(k_{zF\alpha} + k_{zF\beta}) / 2\pi^2 \hbar c, \tag{18}$$

and

$$k_{zF\alpha,\beta} = [2m^* \varepsilon_F (1 + \varepsilon_F/\varepsilon_g) / \hbar^2 \pm m^* g_0 \mu_B H / \hbar^2 - eH/\hbar c]^{1/2}. \tag{19}$$

An iteration procedure was used next to solve the equation

$$\varepsilon_\beta(k_{zF\beta}) - \varepsilon_\alpha(k_{zF\beta}) = \hbar\Delta\omega, \tag{20}$$

$$\varepsilon_{\beta,\alpha} = (\varepsilon_g/2)\{[1 + (2/\varepsilon_g)(\hbar eH/m^* c + \hbar^2 k_z^2/m^* \pm g_0 \mu_B H)]^{1/2} - 1\}. \tag{21}$$

That solution of (20) which corresponds to resonance for the transition 3 was assumed (in the first variant of the calculation) to be the SR position. At low concentrations, i.e., at $\varepsilon_F < \varepsilon_\beta(0) - \varepsilon_\alpha(0)$, the position of the resonance was assumed to be $H_0 = \hbar\Delta\omega / g_0 \mu_B$, as already mentioned above.

A remarkable future in the comparison of the results of such a calculation (the dashed curve in Fig. 2) with experiment is not only the poor quantitative agreement at high Fermi energies, but also the sharp kink of the calculated curve at $\varepsilon_F \approx 24$ meV, which marks the transition from the quantum limit to the concentration region in which the upper Landau subband begins to be filled.

Calculations, by Brueck, Mooradian, and Blum,[5] of the line width of the Raman scattering with spin flip show that it is essential to take into account from the very beginning the electron momentum relaxation, i.e., the motional narrowing of the line. In this case we have not simply replacement of the asymmetric line by a Lorentz line, but a shift of the maximum of the line away from the position corresponding to the minimum k_z towards progressively stronger fields. In the final variant of the computer calculation we therefore used directly the function (15) in place of the resonant function (12).

To describe the observed line shape it was also necessary to take into account the multifrequency operating regime of the CO_2 laser. To this end all the calculations were performed for the five frequencies $\Delta\omega$ of highest intensity, namely 99.1, 100.9, 102.7, 104.5, and 163 cm^{-1}, and the intensities of these lines were normalized in a ratio 0.58:0.95:1:0.95:0.58, obtained by averaging a number of measurements of the SR.

Using expressions (16) and (17), it is possible in the case of weak nonparabolicity to separate in the formula for the power $P(\omega_3)$ the ratio of the resonant and nonresonant parts of the susceptibility:

$$P(\omega_3)\sim(bn)^2\left\{1+2\frac{c}{b}\frac{r'}{n}+\left(\frac{c}{b}\right)^2\left[\left(\frac{r'}{n}\right)^2+\left(\frac{r''}{n}\right)^2\right]\right\}, \tag{22}$$

where $\chi_{nr}=bn$, n are the electron concentrations, $\chi_r=c(r'+r'')$, b and c are constant coefficients,

$$r'=n_t\,\mathrm{Re}\,[\Omega/(1+i\tau_p^{-1}\Omega)], \tag{23}$$

$$r''=n_t\,\mathrm{Im}[\Omega/(1+i\tau_p^{-1}\Omega)], \tag{24}$$

$$n_t=\int(\rho_{0\alpha}-\rho_{0\beta})\,dk_z. \tag{25}$$

The quantities r' and r'' differ from the real and imaginary parts of the resonant formula (15) because of the normalization of the off-diagonal element of the density matrix $\rho'_{\alpha\beta}$ not to the total number of electrons but to the number of electrons capable of executing the transition from band α to band β [see formula (14)]. The first term in the curly brackets of (22) describes the nonresonant background (in relative units), the second describes the interference of the resonant contribution with the nonresonant background (the dispersion contribution) and the third describes the pure resonant part.

The coefficients b and c can be determined from quantum or quasiclassical calculations of $\chi^{(3)}$. Under the experimental conditions, however, the nonresonant background did not agree with its theoretical value, and was much higher, since in most experiments (except the control experiments) the beam with polarization perpendicular to the magnetic field contained radiation of both frequencies, ω_1 as well as ω_2. We therefore determine in the final computer calculations the function

$$2f\frac{r'}{n}+\left(\frac{r'}{n}\right)^2+\left(\frac{r''}{n}\right)^2, \tag{26}$$

where the parameter f, which replaces the theoretical quantity b/c, was used to fit the calculation to the experiment.

The parameters τ_s, τ_p, and f were chosen to describe in the best manner the dependence of the SR position on the Fermi energy of the electrons, the shape of the resonance line, and the dependence of the SR amplitude on the Fermi energy. The presence of three adjustment parameters, however, does not offer great leeway in their choice, since they turn out to be signigicant in different regions of the electron concentrations. In the purest samples, the dispersion contribution to the resonance line is practically nonexistent, nor is the momentum relaxation time significant, while the width of the individual lines of the rotational structure makes it possible to estimate the spin relaxation time τ_s. The parameter f is estimated from the dispersion contribution to the resonant part of the susceptibility $\chi^{(3)}$ for the most strongly doped samples; in addition, the quantity f determines the absolute value of the calculated amplitude of the resonance (in the entire concentration interval). The momentum relaxation time τ_p influences the sharpness of the individual peaks of the rotational structure in the Fermi-energy region where the inhomogeneous broadening takes place (i.e., outside the region of the quantum limit), and influence also the deviation of theoretical plot of the SR position from the straight line corresponding to the transition at $k_z=0$, in the region of the quantum limit (the bending of the plot of the SR position near $\varepsilon_F\approx$ at 24 meV).

To compare the calculated dependence of the SR amplitude on the electron Fermi energy with experiment, the function (26) must, first, be divided by f^2 and, second, it is necessary in addition to take into account the contribution made to the nonresonant background by the bound (valence) electrons in n-InSb, as was done in the case of the CR of the nonlinear optical susceptibility in Ref. 1. Using the correction determined in Ref. 1:

$$\Phi_f/[\Phi_f+2(\Phi_f\Phi_b)^{1/2}+\Phi_b],$$

where $\Phi_F\sim|\chi_f|^2$ is the nonresonant background due to the free electrons and $\Phi_b\sim|\chi_b|^2$ is the nonresonant background of the bound electrons, we can obtain the final value of the resonance amplitude (at a given Fermi energy ε_F), which can then be compared with the experimentally measured quantity.

The results of the calculations are compared with experiment in Figs. 1, 2, and 4. In Fig. 1b (curves 1′, 2′, 3′) the calculated form of the SR lines is compared with experiment (Fig. 1a, curves 1, 2, 3). The calculation accounts satisfactorily for the SR line shape in all three cases: at good resolution of the rotational spectra of the spectrum, strong smearing of the rotational lines, and total broadening of the line with an appreciable contribution of the dispersion part of the resonant component of the susceptibility. The somewhat smaller width of the resonance lines in the case of the calculated curves is due to the fact that only five of the strongest rotational lines were taken into account, whereas a larger number was observed in experiment.

The solid lines in Fig. 2 show the results of the final calculation of the dependence of the SR position on the electron Fermi energy. It is seen that the agreement between calculation of experiment is essentially improved compared with the first variant of the calculation, although at high electron energies some discrepancy between the calculations and experiment still re-

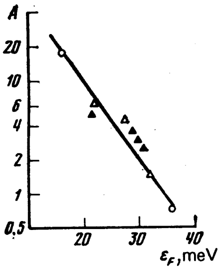

FIG. 4. Dependence of the amplitude of the spin resonance of the nonlinear optical susceptibility $\chi^{(3)}(-\omega_3, \omega_1, \omega_1, -\omega_2)$ on the Fermi energy of the conduction electrons in n-InSb. Points—experiment: Δ – [100] ∥ **H**, ▲ – [111] ∥ **H**, ○—not oriented. Solid curve—calculation.

mains. In this concentration region, however, anomalies are observed in the behavior of the SR, and these are not considered in the present paper.

The dependence of the amplitude of the resonance on the Fermi energy as compared with the data in Fig. 4. It is seen that the agreement is also satisfactory in the Fermi energy region from ~16 to ~35 meV. In samples of higher purity ($\varepsilon_F < 15$ meV) the ratio of the amplitude of the resonance to the nonresonant background was determined with more accuracy (because of the smaller background) and was less accurately obtained in different experiments, possibly because of saturation of the spin resonance.[11]

The best agreement between the calculations and experiments was obtained at the parameter values $\tau_s = 1 \times 10^{-11}$ sec, $\tau_p = 0.85 \times 10^{-12}$ sec, and $f = 0.7$. The momentum relaxation time agrees with the value taken from the average mobility for the given interval of the electron concentrations; the spin relaxation time is noticeably lower than the values obtained from the measurements of the combined resonance[18] and of the SR of the nonlinear susceptibility[6] $\chi^{(2)}(-\omega_3, \omega_1, -\omega_2)$, which were measured however, in samples with lower electron concentrations and in weaker magnetic fields. As to the parameter f, it corresponds to a nonresonant susceptibility exceeding the theoretically expected value by a factor of several dozen, owing to the contribution made to the background by radiation of pure transverse polarization, and also to inexact superposition of the beams with crossed polarization.

We have thus shown in the present paper the theory of nonlinear optical susceptibility constructed with allowance for the band structure of n-InSb and for both the spin and momentum relaxation of the conduction electrons makes it possible to describe satisfactorily the picture the SR of nonlinear optical susceptibility in the wide range of free-electron concentrations.

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