Resonance of spin optical nonlinearity in n-InSb

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We have observed in the present study resonance of the nonlinear optical susceptibility of third order $\chi^3(\omega_3, \omega_1, \omega_1, -\omega_2)$, which is responsible for the frequency shift $(\omega_3 = 2\omega_1 - \omega_2)$, at the spin frequency $\omega_s = g\mu_B H/\hbar = \omega_1 - \omega_2$ of the conduction electrons of n-InSb. This phenomenon constitutes a unique combined resonance, in which the transition of the electrons with spin flip takes place between the Landau sublevels in the magnetic field under the influence of electric fields of laser radiation with frequencies ω_1 and ω_2.

It is known that the nonlinearity connected with the spin flip in n-InSb is large, and has been thoroughly studied in processes of spontaneous and stimulated Raman scattering in a magnetic field,[1,2] and was also observed in the process of generation of far-infrared radiation at $\lambda \approx 110\ \mu$.[3,4]

The nonlinear optical susceptibility $\chi^{(3)}(\omega_3, \omega_1, \omega_1, -\omega_2)$ for the conduction electrons in n-InSb, with allowance for the spin, can be determined by solving the electron equations of motion (which are coupled as a result of the spin-orbit interaction) for the translational and spin degrees of freedom in strong static magnetic field and a strong high-frequency electromagnetic field. These equations are obtained from the Hamiltonian by the effective-mass method described in[5]. The solution of the equations of motion shows that the nonlinear optical susceptibility $\chi^{(3)}$ contains a contribution that has a resonance at the frequency ω_s:

$$\chi^{(sp)}_{xzzx} \sim \frac{\omega_s}{\omega_s^2 - [(\omega_1 - \omega_2) - i\tau_s^{-1}]^2}, \tag{1}$$

where the subscripts x, z, z, x indicate polarization of the radiation at the frequencies ω_3, ω_1, ω_1, and ω_2, respectively, the magnetic field H is directed along the z axis; τ_s is the transverse time of the spin relaxation. The expression for $\chi^{(sp)}$ is written out here for the case of linearly polarized light.

We used in the experiment a Q-switched CO_2 laser generating pulses of 1 kW power at a frequency ω_1 945 cm^{-1} and 200 W at $\omega_2 \approx 1045\ cm^{-1}$, in which case the radiation at ω_3 had a wavelength $\lambda = 11.8\ \mu$. The experimental geometry is shown in Fig. 1. The laser radiation was split into two beams, in each of which radiation of only one frequency was separated, ω_1 or ω_2. According to (1), for the resonance to be observable the radiation with frequency ω_2 should be polarized perpendicular to the magnetic field; to this end, its plane of polarization was rotated through 90° with a half-wave plate. Both beams made congruent and focused on an n-InSb sample placed in a superconducting solenoid. The sample thickness was close to the coherence length $l_c \approx 1.3$ mm and were placed directly in liquid helium at 1.8 °K. The radiation at the bias frequency $\omega_3 = 2\omega_1 - \omega_2$ was separated from the high-power pump radiation by a filter and mono-

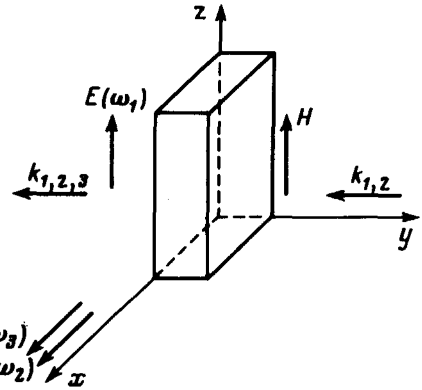

FIG. 1.

chromator, and was registered with a Ge: Hg photoreceiver. The polarization of the radiation at 11.8 μ was determined with a polarizer.

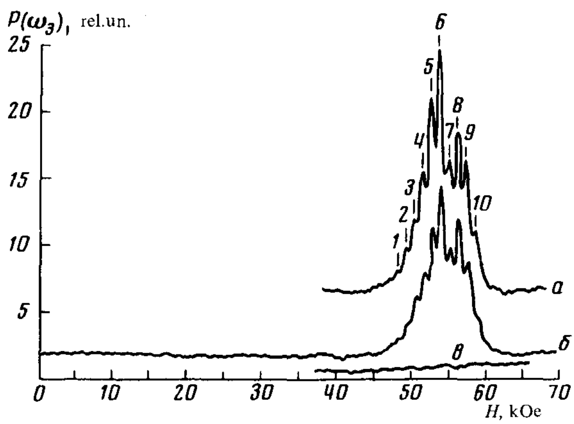

FIG. 2.

Figure 2 shows the dependence of the 11.8-μ radiation power against the magnetic field for two n-InSb filters with conduction-electron densities $n = 2.25 \times 10^{15}$ cm^{-3} (a) and $n = 6.5 \times 10^{15}$ cm^{-3} (b). As seen from Fig. 2, a series of intense not-fully-resolved ω_3 radiation peaks is observed in magnetic fields near 50—60 kOe. Control plots of the signals at the frequency ω_3 show that there are no resonances at $\mathbf{E}(\omega_1) \parallel \mathbf{E}(\omega_2) \parallel \mathbf{H}$ and $\mathbf{E}(\omega_1) \parallel \mathbf{E}(\omega_2) \parallel \mathbf{H}$ in accord with Eq. (1). (One of the control plots is curve c of Fig. 2.) The position of the series of peaks corresponds to resonance at the spin frequency $\omega_s = \omega_1 - \omega_2 \approx 100$ cm^{-1}. The observed series is due to the multifrequency operating regime of the CO_2 laser which generates at several frequencies in the regions of 10.6 and 9.6 μ.

Using the data given in the reviews[6,7] on the spectrum of the generation on the rotational levels of the CO_2 molecule, as well as the energy level scheme calculated by us for the electrons of the conduction band of n-InSb, we can identify all the lines of the observed spectrum (Table).

Numbers of resonance peaks	1	2	3	4	5	6	7	8	9	10
$\omega_1 - \omega_2$, cm^{-1}	91.7	93.6	95.5	97.3	99.1	100.9	102.7	104.5	106.3	108.0
H, kOe	48.4	49.6	50.6	51.9	53.0	54.1	55.4	56.5	57.6	58.7
g-factor	40.5	40.3	40.3	40.0	39.9	39.8	39.6	39.5	39.4	39.5

Our classical calculation of the nonlinear optical susceptibility $\chi^{(3)}(\omega_3, \omega_1, \omega_1, -\omega_2)$ enables us to estimate the ratio of the resonance signal to the monotonic background. This estimate yields a value of approximately 10^4, which is obviously too high, since the classical theory does not take into account the dependence of the resonant component on the occupation numbers of the two spin levels of the Landau ground level. With increasing electron density, the ratio of the resonance signal to the background should increase to a certain maximum value, and then decrease to zero at the concentration at which both spin sublevels of the zero Landau level turn out to be fully occupied. Indeed, resonance was not observed in the n-InSb sample with concentration $n = 4 \times 10^{16}$ cm^{-3}, for which the Fermi energy ϵ_F exceeded the energy $\epsilon_{0,-1/2}$ of the upper spin sublevel, but was observed in the sample with concentration $n = 1.7 \times 10^{17}$ cm^{-3}. In the latter case, the electron transition with spin flip takes place between two sublevels of the Landau level with quantum number $N = 1$.

In the described experiments we were able to obtain a ratio of the resonance signal to the background as high as 100.

The width of the observed resonance lines reaches ~800 Oe, which is much larger than the line width when resonance is observed in the 100 μ region[3] and much larger than the combined-resonance line width.[8] This circumstance may be due to the relatively large absorption of the pump power in the sample, which can lead to a considerable increase of the electron-gas temperature and, as a consequence, to temperature broadening of the resonance lines.

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