Stimulated far-infrared emission from combined cyclotron resonances in germanium

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We have measured the spectrum of stimulated far-infrared emission from Be- and Zn-doped germanium single crystals in crossed electric and magnetic fields. The spectrum consists of broad radiation bands in the frequency range of 40 – 130 cm^{-1} that are due to intervalence band transitions and new narrow emission bands with their center frequencies rising linearly with magnetic field. We demonstrate that the latter emission is due to cyclotron resonance hole transitions between Landau levels of the light-hole subband accompanied by simultaneous spin flip, ‘‘combined resonances.’’
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The possibility of generating coherent far-infrared (FIR) radiation by the inversion of the hot-carrier population in a bulk semiconductor was first considered four decades ago.[1] However, it was only in the 1980’s that FIR lasing was observed from *p*-type germanium single crystals placed in crossed electric and magnetic fields at liquid helium temperatures.[2–4] Stimulated emission can occur through either intervalence band (IVB) transitions between the light-hole (lh) and heavy-hole (hh) subbands or cyclotron resonance (CR) transitions between light-hole Landau levels.[5] Thus far, all spectral investigations have been performed with Ge crystals doped with shallow hydrogenic acceptors (mostly Ga), all of which exhibit internal hole transitions that partially overlap in energy with IVB transitions.[6–10] The interference of dopant absorption and lasing has made the interpretation of the emission spectrum very complicated often leading to speculative assignments of the hole transitions responsible for stimulated emission.

Here we present spectral measurements of the stimulated emission from germanium crystals doped with the double acceptors Be and Zn. Neutral Be and Zn have ionization energies of 25 and 33 meV, respectively, and, therefore, do not lead to self-absorption in the emission range. We have recently demonstrated laser action from Ge crystals doped with these double acceptors[11] and the copper triple acceptor.[12] Here we show that these novel dopants have made possible unambiguous spectral investigations and have led to the discovery of stimulated emission from combined cyclotron resonances.[13]

For this study we used samples from two Czochralski-grown single crystals, one doped with Be and the other with Zn. Lasers were produced from wafers as described elsewhere.[11] The wafers were characterized by variable temperature Hall-effect measurements and photothermal ionization spectroscopy. For both crystals we determined the double acceptor concentration to be 1.5×10^{14} cm^{-3} and a

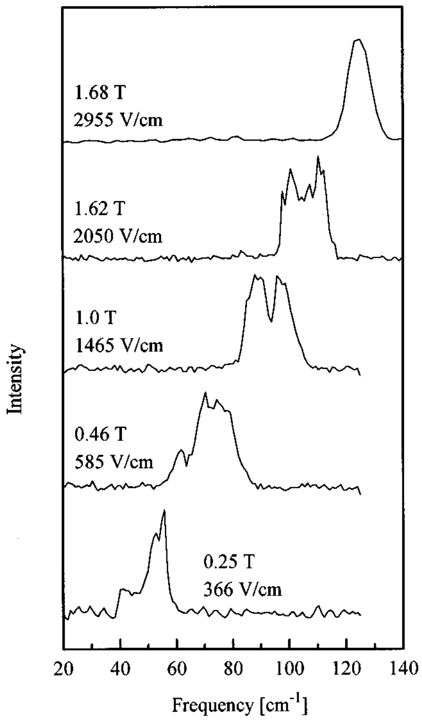

FIG. 1. Stimulated emission spectra for various combinations of E and B fields. The spectral resolution is $1\ \mathrm{cm}^{-1}$ except for the spectrum measured at 1.68 T which has a resolution of $2\ \mathrm{cm}^{-1}$.

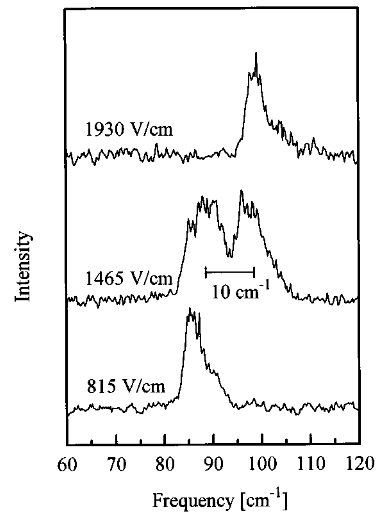

FIG. 2. Stimulated emission spectra at $B=1$ T as a function of electric field (resolution $0.25\ \mathrm{cm}^{-1}$).

residual net shallow acceptor concentration of 1 to $2\times10^{12}\ \mathrm{cm}^{-3}$. The laser samples were mounted between two copper electrodes and placed inside a superconducting magnet in a liquid helium cryostat. Electric field pulses with lengths of 1 to 2 μs were applied. The high refractive index of germanium enables laser operation without external resonators by internal reflection modes. The radiation was detected in the Faraday configuration with the outcoupling direction parallel to the magnetic field. We measured the emission spectra by Fourier transform spectroscopy using a Michelson interferometer and a broadband 4.2 K bolometer.

Figure 1 shows the emission spectra of a $4\times4\times20\ \mathrm{mm}^3$ Ge:Be laser for different combinations of E and B fields. The magnetic field was oriented parallel to the long axis of the crystal, which pointed along a $\langle110\rangle$ crystallographic direction. The electric field was applied perpendicular to B and parallel to a $\langle001\rangle$ direction. By varying both fields we were able to achieve IVB stimulated emission throughout the range of $40-130\ \mathrm{cm}^{-1}$, unlike the case for Ga-doped Ge lasers, which exhibit an emission gap between 60 and $80\ \mathrm{cm}^{-1}$. Figure 2 shows the E-field dependence of the emission at $B=1$ T. Two emission bands are observed. These shift very weakly with E, but the intensity of the higher-frequency band increases with higher field at the cost of its lower-frequency counterpart. Internal absorption by neutral impurities has been excluded as the cause for the signal minimum between the two bands since strong emission at this frequency can be generated at $B=1.16$ T and $E=1.46$ kV/cm.

We have made additional spectral investigations at $E/B=1$ kV/cmT while tuning B up to 3 T. For B higher than 1.5 T a new emission line of relatively small width appears at $78\ \mathrm{cm}^{-1}$ (labeled I in Fig. 3). A slight change in the magnetic induction from 1.5 to 1.55 T results in a shift of the line position to higher energy and in a drastic increase of the line intensity. Upon further increasing the magnetic induction the frequency increases linearly and the line broadens significantly. At $B=2$ T this emission has achieved a broadband character and yet another line structure (labeled II in Fig. 3) appears at $63\ \mathrm{cm}^{-1}$. The frequency positions of lines I and II are shown in Fig. 4. Changing the crystallographic orientation of the outcoupling (and therefore also the B-field orientation) from $\langle110\rangle$ to $\langle100\rangle$ suppresses the narrowband emission I and II. This interesting behavior of the stimulated emission has not been observed for shallow acceptor doped Ge crystals due to dopant self-absorption.

Laser crystals doped with the Zn double acceptor show both the broadband emission that is B-field tunable up to $130\ \mathrm{cm}^{-1}$ and the double band structure observed at 1 T. In addition, the low-frequency lines I and II occur at the same position (Fig. 4). We find that samples doped with the copper triple acceptor exhibit similar spectral characteristics to those shown in Fig. 3. Therefore, the spectral nature of the observed emission must be related to the intrinsic properties of the Ge valence band and is not due to the dopant species.

We attribute the E-field dependence of the broadband emission (Fig. 2) and the line structures shown in Fig. 3 to "combined resonance" hole transitions, i.e., cyclotron resonances between light-hole Landau levels accompanied by a simultaneous spin flip.[13] In the absence of spin flip, cyclotron resonance (CR) emission in Ge is characterized by a line position given by multiples of the cyclotron frequency $\omega_c=eB/m^*$ with a light-hole mass $m^*=0.045\pm0.005m_e$ leading to $\omega_c=20.9\pm2.1\ (\mathrm{cm}^{-1}\ \mathrm{T}^{-1})\times B(\mathrm{T})$. While IVB lasing begins at B fields of about 0.3 T and exhibits broadband emission ($\Delta\omega=20\ \mathrm{cm}^{-1}$), CR emission requires B higher than 1.5 T and is usually characterized by a narrow linewidth of less than $1\ \mathrm{cm}^{-1}$. The occurrence of a simultaneous spin flip means that the hole transitions take place between the so-called "a set" and "b set," which consist of states with $m_j=3/2,-1/2$ and $m_j=1/2,-3/2$, respectively, where m_j is the projection of the total angular momentum along the

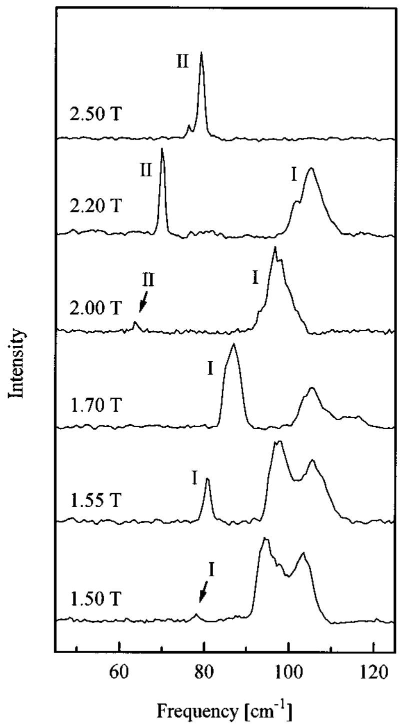

FIG. 3. Stimulated emission spectra as a function of magnetic field (resolution 0.5 cm^{-1}). The ratio E/B is equal to 1.0 kV/cmT for all spectra. The broadband emission at the lower B fields (1.5–1.7 T) is due to intervalence band transitions similar to the spectra in Figs. 1 and 2.

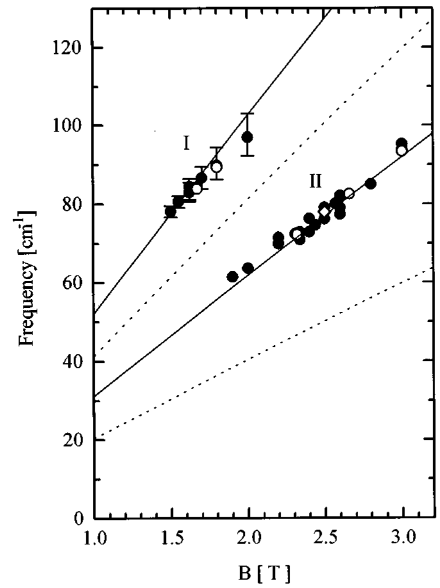

FIG. 4. Frequency dependence of emission I and II with magnetic field. The filled circles, clear circles, and clear diamond represent emission from Be-, Zn-, and Cu-doped germanium laser crystals, respectively. The lines represent the calculated frequencies for the transitions $b_1 \rightarrow b_0$, $b_1 \rightarrow a_0$, $b_2 \rightarrow b_0$, $b_2 \rightarrow a_0$ (from bottom to top). The dashed lines do not involve a spin flip while the solid ones do.

magnetic-field direction.[14–17] Calculations have shown an energy shift between the a- and b-set lh Landau levels of approximately 10 cm^{-1}/T.[15,16,18,19] This is in agreement with our observation of emission (labeled II in Fig. 3) that is of relatively narrow linewidth and that depends linearly on B with $\omega_c \approx 31$ (cm^{-1} T^{-1})$\times B$(T). This corresponds to a typical CR transition (21 cm^{-1}/T) with a simultaneous spin flip (10 cm^{-1}/T). This new result is surprising as a transition between the a and b set would be a violation of selection rules. However, lh-hh mixing of the final state can loosen these rules. While combined resonances have been observed in absorption measurements,[13,20] it has been widely believed that the a set does not support laser action.[16,17,21]

The line that appears at 1.5 T (I in Fig. 3) shows a frequency dependence on B of about 51 cm^{-1}/T before it broadens. This is consistent with a CR transition where $\Delta n = 2$,[15,18] combined with a spin flip of 10 cm^{-1}/T as in the case of line II. The general selection rules for cyclotron resonance of holes in germanium have been determined by Kamimura.[22] Transitions with $\Delta n = 2$ may occur for a magnetic-field direction along [110] as in the case of our experiments. Cyclotron resonance transitions with $\Delta n \neq 1$ have been previously observed in absorption measurements.[13,20]

At a magnetic induction of 2 T the CR line I has broadened significantly achieving IVB character. This magnetic-field dependence of the spectra clearly shows the intimate relationship between IVB and CR emission. The lower level undergoes a transition from a lh-type Landau level to essentially a hh-type Landau level due to increased lh-hh mixing. The distinction between IVB and CR emission is thus just one of the degree of mixing of the final hole state of the transition.[17,18]

In order to lend further quantitative support to the above explanation, we have used the Pidgeon and Brown coupled-band model,[23] which includes both quantum and nonparabolicity effects, to calculate the Landau levels in Ge for a ⟨110⟩ B-field orientation. In accordance with the notation established in previous work,[24,17] we label the Landau levels using the following convention: $-1,0,1,\ldots$ for light-hole states and $1,2,3,\ldots$ for heavy-hole states. The applied electric field influences the degree of mixing between the heavy- and light-hole states. For $B<1.5$ T the lh Landau-level energy and spacing are nearly constant for electric fields up to 2 kV/cm.[24] For $B>2.5$ T the Landau-level spacing is still conserved even though the lh levels mix strongly with hh states.[17] We therefore conclude that the transition energies between lh Landau levels for $E>0$ can be roughly estimated by the values at $E=0$.

Calculating the energies of the allowed transitions for $1<B<3$ T and comparing these to the measured spectra, we attribute the line features I and II (Fig. 3) to transitions from the b set to the a set, that is, combined resonances as described above. Line I is assigned to a light-hole transition from the b-set level 2 to a-set level 0 ($b_2 \rightarrow a_0$) while line II is tentatively assigned to the light-hole transition $b_1 \rightarrow a_0$. Calculated energies for various transitions are depicted by the lines in Fig. 4. The data are clearly best described by combined resonances. A double-line structure associated with emission II for certain combinations of E and B suggests the occurrence of transitions involving levels of higher n (e.g., $b_2 \rightarrow a_1$).

The b-set light-hole levels 1 and 2 are the same as those previously assigned to the initial states for CR emission from Ga-doped Ge lasers ($b_2 \rightarrow b_1$ and $b_1 \rightarrow b_0$).[24,17] The difference in the final states may result from the differing direction of the applied electric field in combination with dopant self-absorption present in shallow acceptor doped laser crystals. CR emission from Ga-doped Ge lasers is normally achieved with an applied E field close to a $\langle 110 \rangle$ direction (compared to a $\langle 001 \rangle$ orientation in this study). Changing the direction of E redistributes the hole population among these levels. Inversion is, therefore, achieved between different levels as reflected by the above assignments. The significance of dopant self-absorption is reflected in the absence of combined cyclotron resonance emission from a Ge: Ga crystal that we tested with E and B parallel to $\langle 001 \rangle$ and $\langle 110 \rangle$, respectively, for $E/B = 1$ kV/cmT.

Combined resonance transitions can help explain the double-band structure that is characteristic of the IVB emission spectra of these Be- and Zn-doped Ge lasers shown in Fig. 2. At $B = 1$ T the splitting between the two bands is approximately 10 cm^{-1}. We observed this type of splitting increasing at a rate of approximately 10 cm^{-1}/T for B ranging from 0.75 to 2.2 T. This is consistent with emission involving a spin flip. Increasing the E field while holding B constant increases the degree of mixing of levels in the b set and, therefore, favors emission with spin flip over that without. Our calculations show the IVB emission at $B = 1$ T to result from transitions with $\Delta n > 3$. The lower-energy band corresponds to transitions within the b set while the higher-energy band involves b- to a-set transitions.

In conclusion, double acceptor doped germanium is a model system for the investigation of stimulated emission from hot carriers in semiconductors. By eliminating the effect of dopant self-absorption, we have been able to obtain convincing evidence that both the a-set and b-set Landau levels can participate in the stimulated emission process. In addition, we have shown that the differing spectral character between intervalence band and cyclotron resonance lasing is merely a signature of the degree of mixing between the heavy-hole and light-hole states into which the holes decay radiatively. The results presented here should clear the path for further theoretical developments in the field and encourage continued efforts toward the realization of stimulated emission from other semiconductors such as boron-doped silicon.

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