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Optically detected impact-ionization-related chaotic oscillations in $Ga_{0.47}In_{0.53}As$

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Studies of impact-ionization-induced chaotic oscillations of photoluminescence (PL) intensity by the optically detected cyclotron resonance method are reported in $Ga_{0.47}In_{0.53}As$ layers grown by liquid-phase epitaxy on semi-insulating InP:Fe. It is directly shown that the impact ionization from shallow impurities is responsible for the chaotic oscillations of the PL intensity and the corresponding oscillations in the density and energy of free electrons.

Spontaneous oscillations of the electric current in semiconductors, leading to chaos, have been observed in many materials.[1] Previous theoretical work suggests that chaotic oscillations can be related to impact ionization from impurity levels or excitons.[2] These oscillations appear in electrical measurements at just below or above the threshold field for impurity and exciton breakdown, respectively.[2,3] Very recently oscillations of the carrier density have been observed during impact ionization of excitons in silicon by microwave-field-heated free carriers.[4] The accompanying temporal changes of the photoluminescence (PL) intensity during current oscillations have not been observed up to now, however. In this paper we present experimental evidence of such chaotic PL intensity oscillations. They are detected using the optically detected cyclotron resonance (ODCR) technique and provide a solid ground for the proposed impact-ionization model.

The ODCR technique was presented by Baranov *et al.*[5] and later used by Romestain and Weisbuch.[6] In this experiment a cyclotron resonance (CR) is detected via observation of changes in the intensity of a relevant PL emission. Among various mechanisms responsible for such observations the impact-ionization mechanism turned out to be one of the most efficient.[7] CR is detected due to impact ionization of impurities or of excitons via microwave-field-heated free carriers. Thus, at the resonance condition, when free carriers absorb most efficiently, the PL transitions are affected. This is observed as a decrease in the PL intensity of some bands, whereas other transitions less affected by the impact process may be enhanced.[7] For doped samples the CR spectrum is usually not well resolved, and a broad "nonresonant" signal is then observed.[8] A broad ODCR signal of the conduction-band electrons in $Ga_{0.47}In_{0.53}As$ was observed previously by Kana'ah *et al.*[9] In this study we have employed the ODCR to monitor the impact ionization and then the chaotic oscillations of the PL intensity. We show that these oscillations occur for microwave fields close to the threshold values for impact ionization of shallow donors in $Ga_{0.47}In_{0.53}As$.

The high-purity 2-μm $Ga_{0.47}In_{0.53}As$ layers were grown by the liquid-phase-epitaxy (LPE) technique. They were crystallized by a step-cooling method at 600 °C,[10] on semi-insulating InP:Fe substrates with a 1.5-μm *n*-type (10^{17} cm^{-3}) InP buffer layer. The lattice mismatch between the $Ga_{0.47}In_{0.53}As$ layer and the InP substrate was $< 10^{-4}$. The residual donor concentration was gettered by adding to the melt a small amount of Yb.[11] The obtained $Ga_{0.47}In_{0.53}As$ samples were *n* type with a free-carrier concentration at room temperature of 2×10^{15} cm^{-3}. A description of the experimental setup can be found elsewhere.[7]

In Fig. 1(a) we show the PL spectrum measured at 4 K under above band-gap excitation. This spectrum consists of two unresolved PL bands at 1531 nm (810 meV) and 1562 nm (794 meV), respectively, with a low-energy wing extending up to 1650 nm. The first PL band is attributed to unresolved bound exciton (BE) recombination (due to overlapping of several donor and acceptor BE spectra[12] and alloy broadening), whereas the second band was attributed to band-acceptor (BA) and donor-acceptor pair (DAP) transitions in the previous optically detected magnetic resonance[9] (ODMR) and PL (Refs. 12–14) studies. They had the maxima at 0.801 and 0.779 meV, respectively.[9] (These previous ODMR studies were performed on $Ga_xIn_{1-x}As$ samples obtained by the atmospheric-pressure metal-organic chemical-vapor-deposition method.) In our studies we prove that the 794-meV band consists of two overlapping bands that are of a different nature than proposed previously. We could resolve these bands in the ODCR experiment, as will be discussed in detail below.

The PL spectrum measured with the maximal microwave power available in our experiment is quite different from that shown in Fig. 1(a). The higher energy bands are now totally quenched and a PL subband at 1572 nm (789 meV) dominates [Fig. 1(b)]. Such a PL spectrum was reported previously and was attributed to Si acceptor transitions.[12] The spectrum shown in Fig. 1(b) was measured with 200-mW microwave power and the magnetic

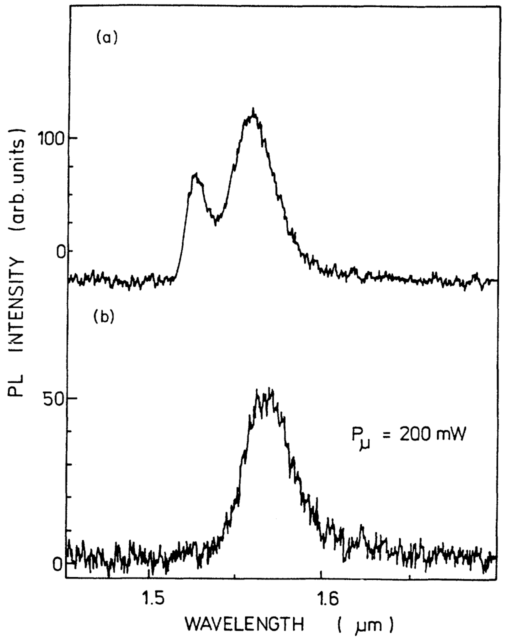

FIG. 1. (a) Photoluminescence spectrum measured at 4 K under 514.5 nm excitation. The spectrum shown in (b) was measured for a magnetic field set at 0.5 T and an applied microwave power of 200 mW.

field set at 0.5 T. The 0.5-T field was selected arbitrarily from the broad CR signal. Since in the experiment shown in Fig. 1(b) the microwaves were not chopped, the PL intensity changes could be dominated by not only the impact ionization, as studied further on, but as well by the bolometric effects.[7,8] (By the bolometric effect we mean here the change of PL due to a rise of the sample temperature.) We have, however, an independent indication that lattice heating by the microwaves is rather small. The 1531- and 1562-nm PL bands should shift to higher energy with a rise in temperature.[12] This is not observed in our experiment. The PL bands are quenched under application of the microwaves, indicating a high efficiency of the impact-ionization mechanism. This effect can be monitored directly by measuring the PL intensity changes in phase with the chopped microwave power. A high chopping frequency (6.25 kHz) minimizes the sample heating by microwaves and allows to separate the effects due to the impact ionization and sample heating.[7,8] The spectral and the microwave power dependencies of the PL intensity are shown in Figs. 2 and 3, respectively. These data show clearly that the 1562-nm band consists of two superimposed bands at 1555 and about 1572 nm.

The data shown in Fig. 3 are consistent with the impact-ionization mechanism for the PL intensity changes. A characteristic threshold dependence is observed. First, the BE band is quenched (above 50 μW), whereas the two superimposed bands at 1562 nm are enhanced. This effect is easy to explain. Free carriers impact ionized from BE's are recombining via channels still not affected directly by microwaves. Above 12.6 mW a distinct change of the PL spectral response to microwaves is obtained. The two

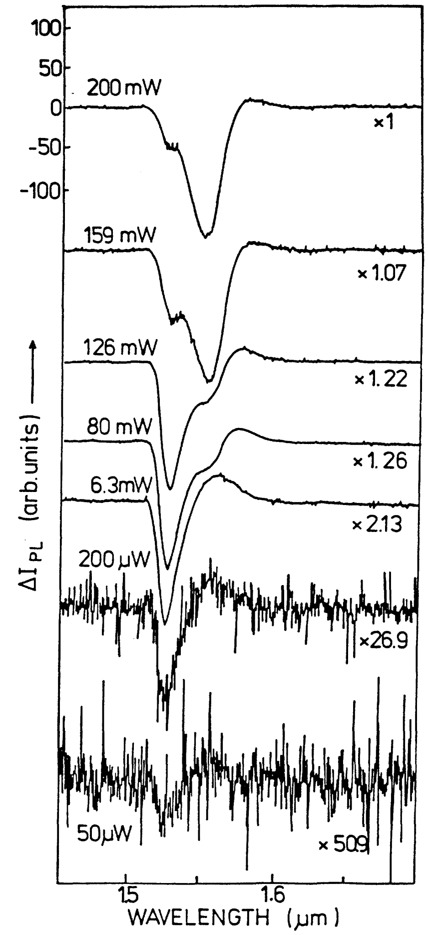

FIG. 2. The spectral dependence of the PL intensity measured at 4 K at different microwave power levels in phase with microwaves chopped at 6.25 kHz.

components of the 1562-nm band now respond differently to the microwaves. The enhancement of the 1555-nm band saturates first and then this band starts to decrease. The 1572-nm band is also affected for a microwave power above 25 mW. This is observed first as a saturation of its magnitude and then as a small decay in its intensity, as shown in Fig. 3. We conclude thus, that the threshold for the 1572-nm PL impact is about 25-mW microwave power.

The observed microwave power dependence of the 1562-nm PL indicates that this band is of a different origin than proposed previously. It was ascribed to either BA and/or DAP transitions.[12] We prove here directly that this PL band is composed of two subbands with different thresholds for impact ionization. With rise in microwave power the higher energy band is reduced first, which is inconsistent with BA and DAP character of the 1555- and 1572-nm bands, respectively. The former should shift to higher energy and should be enhanced when the lower energy DAP band is reduced. We tentatively attribute these PL transitions to DAP processes involving two different donors (of different ionization energies) and, likely, the same type of acceptor.

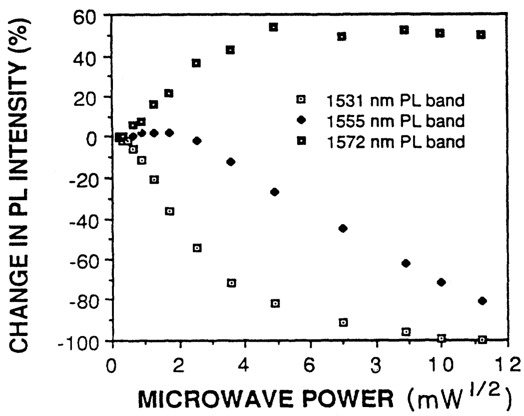

FIG. 3. The relative changes of the 1531-, 1555-, and 1572-nm PL bands vs the square root of the applied microwave power chopped at 6.25 kHz.

A noteworthy effect is observed at further increased excitation intensity. The data shown further on were taken at a laser power increased by 15%. The laser power dependence of this effect was very nonlinear. By this effect we mean irregular PL intensity oscillations, as shown in Fig. 4(a). These oscillations were so large that they also could be observed directly in the PL intensity. It was, however, easier to observe them in the ODCR experiment, which is directly sensitive to any microwave-induced PL intensity changes. The irregular oscillations were observed with low repetition frequency of up to 6.5 s [Fig. 4(b)]. The amplitude and period of these oscillations rise rapidly with an increase in the microwave power applied. They were not observed for the microwave power below 25 mW, i.e., the threshold for 1572-nm PL impact ionization.

All the features of these oscillations agree with the impact-ionization PL breakdown. We observe that the chaotic oscillations occur at a microwave field just above that for the 1572-nm band breakdown. It is the feature expected for the impact-ionization driven force for the chaos.[2] The following explanation can be proposed to account for the experimental results. This explanation is similar to the one proposed previously by Schöll and coworkers[15,16] for self-generated chaotic current or voltage oscillations. In ODCR experiments free carriers are heated by the microwave power. First they reach an energy sufficient for impact-ionizing donor and acceptor BE's. Electrons and holes impact ionized from BE's are recaptured immediately by donor and acceptor centers. This is observed as an enhancement of the superimposed 1562-nm PL band. At an increased microwave power the accelerated electrons can impact ionize the shallower donors. The "hydrogenic" donor ionization energy in $Ga_xIn_{1-x}As$ is only about 2.6–3 meV and is much smaller than the shallow acceptor ionization energy. Thus, at increased microwave power donors are ionized first. This is observed as a decrease of the higher energy DAP transition, but the 1572-nm emission is still enhanced. Once the deeper donors are impact ionized the free-electron concentration rises. Their energy is, however, reduced rapidly by

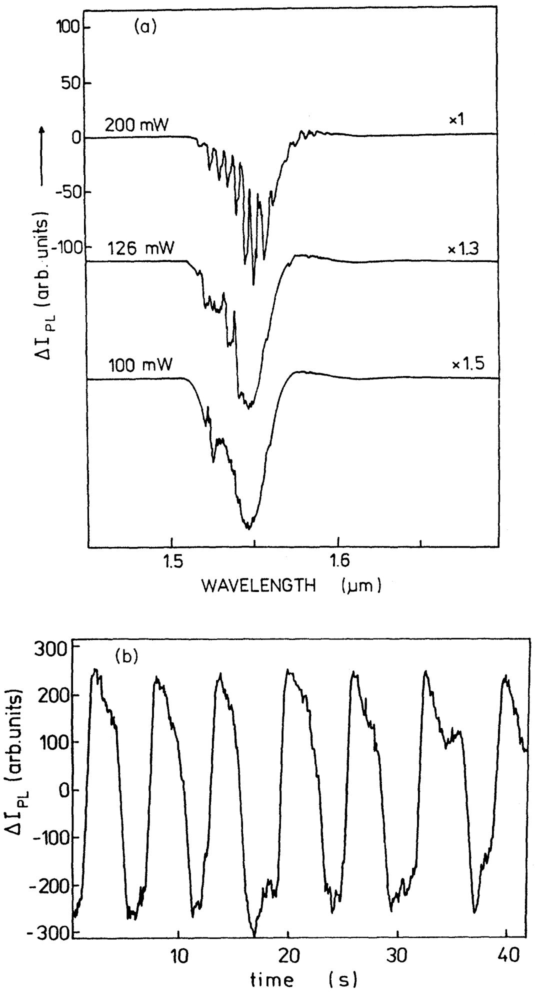

FIG. 4. (a) The temporal changes of the PL intensity measured at increased (by 15%) excitation intensity and 200-, 126-, and 100-mW applied microwave power. In (b) the time dependence of the chaotic PL intensity oscillations is shown, with the detection set at the resolved 1572-nm band and for 200 mW of applied microwave power.

the impact ionization of donors and the consequently enhanced electron-ionized donor and electron-phonon scattering. The reduction of electron energy results in their rapid recapture, observed in our experiment as a rise in the PL intensity. Once part of the free electrons is recaptured by the donors, the carrier scattering is reduced due to decreased free-electron concentration and a reduced number of the ionized donors. Thus, the microwave-induced electron heating becomes more efficient. This in turn results in a reduction in PL intensity due to the impact-ionization mechanism, which closes the cycle. The PL intensity starts to oscillate. The oscillation period depends on the frequency of the driving force (microwave chopping frequency) and on the cooling and heating times of the electron gas. The amplitude of the resulting intensity oscillations is the largest for those emissions that are the most sensitive to the impact ionization. The fact that oscillations are observed on the whole PL spectrum, but with different amplitude is, thus, consistent with the above explanation of the experimental results. "Cooled" carriers can be recaptured into BE's and the shallower donors which are, otherwise, totally or partly (donors) impact ionized. The oscillations are smallest for the 1572-nm PL band.

The oscillation rate (which is irregular) depends on the free-electron concentration and their average temperature, which in turn depends on excitation intensity and microwave power, and on microwave chopping frequency. We could not follow these dependencies in detail since they are very nonlinear. Once the excitation intensity is increased, the free-electron concentration rises and the carrier scattering is changed. Then the microwave power must be increased to reach the threshold for the impact ionization. Thus, all the experiments presented were performed for similar laser excitation intensity, which was necessary to compare the threshold for the 1572-nm PL impact and the PL intensity oscillations. We found that these thresholds differ only slightly due to the above mentioned dependence on free-carrier concentration (excitation intensity). This is an important observation since we can prove that the observed oscillations are due to the impact ionization of the excitons and shallow donors, i.e., due to changes in free-electron concentration and kinetic energy.

Concluding, we show here the experimental observation of chaotic oscillations of the PL intensity that accompany the oscillations in the free-carrier density in a semiconductor. We prove directly that the impurity impact ionization is a driving force to chaos. The observed oscillations are accounted for by corresponding oscillations in the average free-carrier kinetic energy. These optical observations are important complements to previous electrical measurements, and put the impact-ionization model as the driving force for chaos on a firm experimental ground.

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