Ionization of the Direct-Gap Exciton in Photoexcited Germanium

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The excitonic absorption at the direct gap in Ge is measured as a function of the density and temperature of excitations at the indirect gap. The spectra are analyzed by means of a many-body theory which is valid for arbitrary free-carrier densities. The critical densities for the ionization are determined for temperatures from 8.5 to 60 K and compared with other predictions.

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In photoexcited semiconductors a transition from an insulating state to a conducting state occurs[1] with increasing excitation intensity. This transition is due to the screening of the attractive Coulomb potential between an electron (e) and a hole (h)[1-2] by the other bound- and free-pair excitations. A consistent treatment of the contribution of both the excitons and the free carriers to the screening for finite temperatures and arbitrary excitation intensities does not exist. The first steps toward this goal have been done by Zimmermann,[3] Silin,[4] Bisti and Silin,[4] Comte and Nozières,[5] and Röpke, Meister, Kollmorgen, and Kraeft.[6] It has been shown that excitons alone are much less efficient in their screening action than free carriers because of the presence of a gap of about one exciton rydberg. The concentration of excitons needed to ionize an exciton is according to Ref. 4 about 50 times larger than the corresponding concentration of free carriers. Recent picosecond experiments by Ulbrich and coworkers[7] confirm that the ionization concentration decreases strongly if the exciting beam is tuned away from the exciton into the band-to-band absorption region.

Therefore, under experimental conditions where free carriers are available as a result of elevated temperatures of nonresonant excitation, the screening is dominated by these free carriers. In the present paper we will present a rather detailed investigation of the exciton ionization in Ge performed by measuring with a weak test beam the absorption spectrum of the direct gap while a stronger cw-pump beam generates e-h pairs which relax into the indirect-gap minima. We will show that the obtained spectra can be described excellently with only minor deviations by use of a recently developed microscopic theory[8,9] which incorporates the screening due to an arbitrary concentration of free carriers. The simple Mott criterion $k_0 a_0 = 1.19$, where k_0 and a_0 are the inverse Debye screening length and the exciton Bohr radius, indicates that the exciton binding energy vanishes at densities where the free-carrier plasma is still nondegenerate. Under this condition the emission line shape of the plasma is merely a function of the temperature, but is independent of its density. Therefore, absorption spectra[10] are better suited than luminescence spectra[11-14] for the investigation of the exciton ionization. As will be shown in this paper, it is not possible to use a temperature-independent scaling factor[13] between the excitation power and the carrier density in order to determine the transition densities.

The study of the exciton ionization by measurement of the absorption of the direct gap in Ge, especially, has the advantage that band-filling and exchange effects in the conduction band are absent, because the screening electrons are at the indirect-gap minima of the L point. Additionally, the nonlinearities which may result if the observed screened pairs are identical with the screening pairs are suppressed. But, most important, very homogeneous densities in space and time can be realized, because the long lifetimes of the excited carriers permit the use of continuous excitation with an unfocused laser beam. Therefore, absorption experiments at the direct gap in Ge are especially well suited to test the theoretical description of the ionization process. It should be mentioned that the study of an exciton interacting with a homogeneous plasma has to be performed above the critical temperature for droplet formation.[12] The droplet condensation causes a spatial inhomogeneity and prevents one

from increasing the carrier density continuously up to the critical Mott density.[15]

For our measurements we used high-purity Ge single-crystal samples. In plane-parallel polished platelets thin windows were etched with typically 2 mm diameter and 10 μm thickness. The thickness was controlled by the Fabry-Perot interferences in transmission measurements. To ensure a high homogeneity of the electron-hole plasma in the thin observation region, we excited the sample from the rear side of the platelet with an unfocused Nd-doped yttrium aluminum garnet laser in cw operation. The transmitted light from a projector lamp was dispersed by a 1-m grating monochromator, detected by a PbS photoresistor, and processed by a lock-in amplifier. The spectral range of the probe lamp was limited to energies between 878 and 898 meV by a further monochromator in order to avoid additional excitations. The data were corrected for the detector and apparatus sensitivity functions. The absorption spectra have been measured for lattice temperatures T_L between 8.5 K and 60 K. Simultaneously with the transmission, we measured the luminescence due to the L-point electrons which provided independent information on the carrier temperatures.

Figure 1 shows the measured spectra at $T_L = 30$ K for excitation intensities of $I = 0$, 10, 30, and 150 mW. With increasing excitation the excitonic absorption continuously disappears, indicating the increasing ionization of the bound state.[16] In order to determine the free-carrier densities

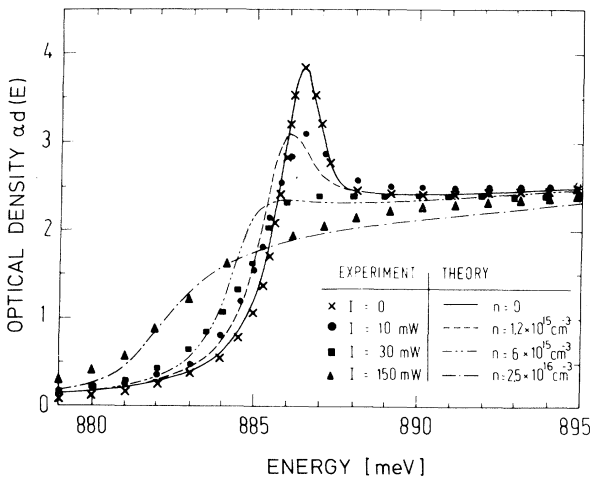

FIG. 1. Measured and calculated absorption spectra at the direct gap of Ge at a lattice temperature $T_L = 30$ K for various excitation intensities I.

of the different spectra the experimental data are analyzed with use of a recently developed many-body theory.[8,9] The calculations of the complex optical dielectric function are based on the solution of the Bethe-Salpeter equation in k space by matrix inversion. The theory incorporates consistently the screening and renormalization effects by free carriers, but neglects the corresponding exciton contributions. Thus, the theory is appropriate for the high-temperature range where a sufficient concentration of ionized pairs exists. Screening and self energies are calculated in a quasistatic modified plasmon-pole approximation, in which dynamical effects are incorporated. Similar quasistatic self-energy calculations have also been used recently in related theoretical descriptions.[5,17] Note that the random phase approximation (RPA) and its plasmon-pole approximation yield, at higher temperatures (to which our theory is limited), a valid low-density limit, which is characterized by the Debye-Hücke screening. This is different from the situation at low temperatures, where RPA is only justified for high densities, i.e., $r_s \ll 1$.

For the calculations the following material parameters have been used: $E_g(\Gamma) = 888.4$ meV; binding energy of the Γ exciton $E_B{}^x = 1.9$ meV; $\epsilon_\infty = 16.8$; $\epsilon_0 = 15.3$; damping of the pair state $\gamma = 0.48$ excitonic rydberg; electron mass at the Γ point $m_e{}^\Gamma = 0.037 m_0$, where m_0 is the free-electron mass. For the values of the density-of-states masses and optical masses see Beni and Rice.[18]

Once the zero-density spectrum is fitted by adjustment of the damping constant γ, which for simplicity is assumed to be density independent, the theory contains no further adjustable parameters, neither for the *amplitudes* nor for the *energetic positions* of the spectra. In the complete temperature and density range the experimental and calculated shapes of the absorption spectra are in excellent agreement as shown in Fig. 1 at 30 K. The same holds for the energetic positions at densities below and above the ionization of the exciton. Close to the ionization density a small discrepancy (< 0.3 Ry) in the form of a slight red shift of the exciton occurs. This red shift in the calculated spectra is due to small inaccuracies caused by the approximations involved in the self-energy calculation, so that the compensation between the reduction of the exciton binding energy and the band-gap shift is not perfect. Outside the ionization regime the discrepancy is much smaller as can be seen in Fig. 2 where the exciton

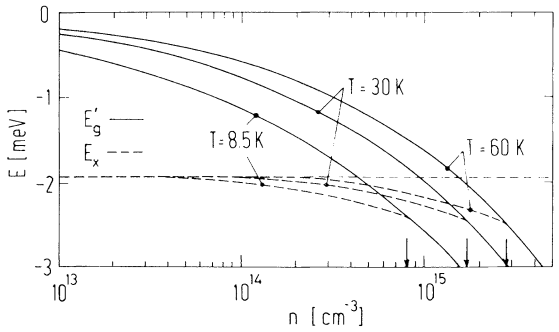

FIG. 2. Calculated values of the renormalized energy gap E_g' and of the exciton energy E_x as function of the free-carrier density n. The dashed line depicts the experimentally observed exciton position.

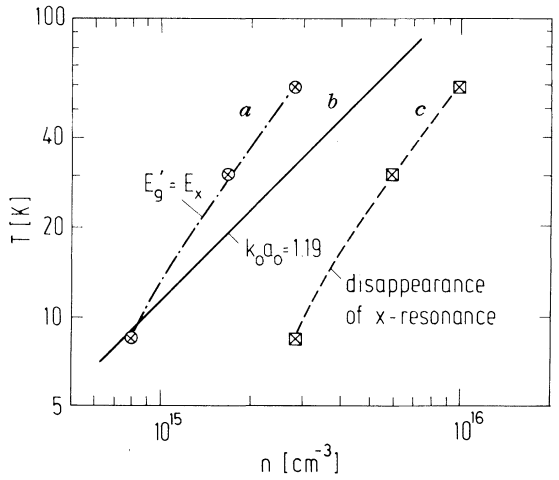

FIG. 3. Temperature vs free-carrier density for different ionization criteria: curve a, $E_x = E_g'$; curve b, $k_0a_0 = 1.19$, and curve c, from the absorption spectra at the disappearance of the x resonance.

binding energy and the renormalized gap are plotted as functions of the free-carrier density.

From a comparison of the shapes of the experimental and theoretical spectra as shown in Fig. 1 a precise evaluation of the free-carrier densities is possible. As mentioned earlier, the fraction of the bound e-h pairs will give a much smaller contribution to the screening and is therefore disregarded. Note that this procedure allows for the first time a reliable determination of the low free-carrier densities, which cause the exciton ionization. The resulting free-carrier densities for the spectra depicted in Fig. 1 are $n = 0$, 1.2×10^{15} cm^{-3}, 6×10^{15} cm^{-3}, and 2.5×10^{16} cm^{-3}. The curve for a density of 1.2×10^{15} cm^{-3} is close to the exciton ionization density, for which the binding energy vanishes (see Fig. 2).

Because of the Coulomb enhancement the absorption spectra show strong deviations from the simple square-root energy dependence even above the ionization density (Fig. 1). One might be tempted to identify the "flat" spectrum at a density of 6×10^{15} cm^{-3} with the onset of ionization, because the "exciton" resonance has just disappeared. However, the theory indicated that this phenomenological criterion overestimates the Mott density by a factor of about 4. In Fig. 3 we compare various ionization criteria. The curve $n(T)$, which is obtained from our criterion of the ionization ($E_x = E_g'$, curve a) is in close agreement with a result obtained by Zimmermann *et al.*[19] using a low-density limit theory. The static Mott criterion (curve b) yields somewhat higher densities than the criterion $E_x = E_g'$. The "flat" spectra give the highest densities (see curve c), with a temperature dependence very similar to that of the actual ionization curve a.

We want to emphasize that the experimental excitation intensities cannot be used for estimates of the ionization density. The intensities necessary to obtain "flat" spectra (Fig. 3, curve c) decrease with increasing temperature. This is due to the temperature dependence of the ionization degree of the excitations at the indirect gap and of their diffusion rate to the surface of the very thin samples.

In conclusion, the first quantitative analysis of the ionization process of the direct-gap exciton in Ge is reported. The experimental findings are analyzed with a theoretical description based on a simplified RPA free-carrier screening which is valid at elevated temperatures in the whole density range. In the investigated temperature and density range no indications of the necessity to include exciton contributions to the screening are found. The observed transition from the exciton to the plasma is very smooth, as expected, because for the present situation the vanishing of the bound state does not give rise to a strong catastrophic increase of the free-carrier concentration.

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$$V_{ij}(q)=V(q)\left[1-\frac{\omega_{pl}^{2}}{2\omega_q}\left(\frac{1}{\omega_q+q^2/2m_i}+\frac{1}{\omega_q+q^2/2m_j}\right)\right],$$

where $i, j=e,h$. This modified plasmon-pole approximation has a similar structure to the Haken potential describing the screening by LO phonons [see, e.g., H. Haken, in *Polarons and Excitons*, edited by C. G. Kuper and G. D. Whitfield (Oliver and Boyd, Edinburgh, 1963), p. 205].

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[16]A qualitatively quite similar evolution of the transient absorption in GaAs has been reported by Shank *et al.* (Ref. 10) from experiments using subpicosecond pulses. Because of the short carrier lifetimes in GaAs excitation intensities ranging from 1 kW/cm^2 to 1 MW/cm^2 are required to ionize the free excitons. For Ge, in contrast, excitation intensities of less than 1 W/cm^2 are sufficient. Hence in Ge the exciton-plasma interaction is studied under quasi thermodynamical equilibrium conditions, whereas in the picosecond data from GaAs nonequilibrium effects as, e.g., carrier thermalization are important.

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