Submillimeter Absorption Spectra and Phase Transition of Indirect Excitons in Germanium

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Abstract—**The results are presented of the detailed studies on the exciton absorption spectra in ultrapure Ge in the spectrum regions 0.67–1.1 and 1.7–3.7 meV with a backward-wave tube radiation. It is shown that the measured spectrum corresponds to the exciton's transitions from the ground to excited states. As a result of temperature measurements at 1.6–4.2 K the phase transition of excitons into the condensed state has been observed without the formation of excitonic molecules.**

I. INTRODUCTION

THE LOWEST excited states in a semiconductor, as is well known, correspond to the crystal excitation, which is accompanied by electron transition from valence into conduction band and formation of hydrogen-like electron-hole systems named excitons. From the very time of the discovery of this phenomenon [1], numerous exciton investigations have been carried out, but exclusively in the region of interband transitions. That means that actually the subject of the studies was the processes of exciton generation rather than the excitons themselves. Meanwhile, it was obvious that more detailed information concerning the energy spectrum and the other properties of excitons can be obtained from the investigations in a spectral region corresponding to the exciton binding energy, i.e., for most semiconductors in the far infrared. For the first time such experiments were performed on germanium with the help of a diffraction spectrometer [2]. The observed absorption in the region 2–5 meV was explained as the photoexcitation and photoionization of the excitons in a crystal. Further progress in the exciton studies was achieved with the help of a more sensitive high-resolution method using a backward-wave tube (BWT) as the monochromatic source of submillimeter radiation [3]–[7].

We present here the results of detailed studies of the absorption spectra of indirect excitons in germanium at 1.5–4.2 K, measured using BWT as the monochromatic source of submillimeter radiation in the energy range 0.67–1.1 meV and 1.7–3.7 meV, and using the diffraction spectrometer in the region 1.8–20 meV. The ultrapure germanium samples used in these experiments had a residual-impurity concentration not exceeding 5×10^{10} cm^{-3} [8] that completely excluded possible impurity effects. Nonequilibrium carriers in the sample (thickness ~ 1 mm) placed into liquid helium, were generated by means of radiation from an incandescent lamp passing through the glass filter and focused on the sample. The mean lifetime of carriers, bounded into excitons at these temperatures, being 5–10 μs, such excitation provided the average volume concentration of excitons $\sim 10^{13}$ cm^{-3}. The far-infrared transmission change in the sample was directly measured due to optically generated excitons. The differential technique with 500-Hz modulation of the intensity of exciting light was used to detect these very small transmission changes. As is well known in this case, the alternative signal is proportional to the optically induced absorption as well as to the intensity of BWT radiation. The use of a BWT as a powerful source of submillimeter radiation made it possible to perform the highly sensitive measurements. The submillimeter output power in the process of BWT tuning was held constant by means of the two-strip grating attenuator. An n-InSb detector, placed at the bottom of waveguide in a helium cryostat, was used to measure the differential absorption signal as well as to control the power level of BWT radiation (chopped with 9 Hz). In order to eliminate interference effects arising from the high coherence of BWT radiation, all details of the waveguide were made from high-absorptive materials as soon as possible.

Manuscript received June 19, 1974.
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II. EXCITON ABSORPTION SPECTRA

The results of a detailed investigation of submillimeter absorption in germanium stimulated by the generated nonequilibrium carriers in a crystal, measured at 4.2 K and 2.0 K with the aid of the differential method described above, are shown in Fig. 1. The measured spectrum can be seen to consist of a complicated system of the absorption lines with a width on the order 0.05–0.1 meV (see, e.g., the lines at 2.875, 3.14, and 3.42 meV) overlapping in certain spectral regions. The positions of the individual lines and absorption maxima, measured at 4.2 K with accuracy not less than 0.01 meV, correspond to the energies: 2.22, 2.35, 2.41, 2.47, 2.52, 2.875, 2.96, 3.03, 3.14, 3.30, 3.35, 3.42, and 3.50 meV. The observed spectrum cannot be explained by the interaction of the submillimeter BWT radiation with the free carriers or with the impurities in a crystal. Since the measured spectrum corresponds to the energy range close to the exciton binding energy, estimated from the interband transition experiments, and consists of a series of narrow lines ($\Delta h\nu < kT$), one can conclude that it is caused by transitions from the ground into excited states of excitons. From Fig. 1 it is obvious that the binding energy of indirect exciton in germanium is more than 3.7 meV.

As can be seen from Fig. 1, the decreasing of tempera-

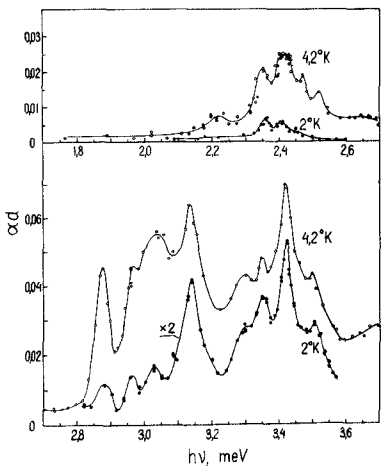

Fig. 1. Spectrum of exciton absorption in ultrapure germanium.

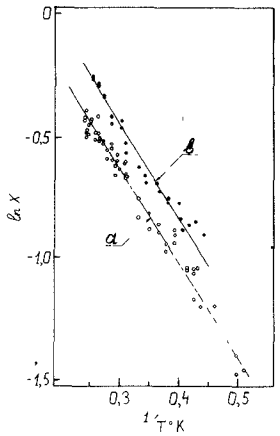

Fig. 3. The temperature dependence of the ratio $x = \alpha_2 d/\alpha_1 d$ [$\alpha_1 d$, at 3.42 meV; $\alpha_2 d$, at 2.88 meV (a) and at 3.03 meV (b); d, the sample thickness].

ture results in a considerable change of intensity of the exciton absorption spectrum. Some of the absorption lines are not very much decreased, while the lines in the other part of the spectrum (see, e.g., 2.88–3.03 meV) have almost completely disappeared. The results of more detailed measurements of the temperature dependence of exciton absorption at different parts of the spectrum are given in Fig. 2. One can see that in the temperature interval 2.5–3.5 K the intensities of the lines at 2.41, 2.45, 3.14, and 3.42 meV change very slightly while the intensities of lines at 2.88 and 3.03 meV drop along with the temperature. It seems natural to assume these two groups of lines correspond to the exciton transitions from the two 1s-type lowest levels of the ground states of the exciton in germanium. Fig. 3 demonstrates the temperature dependence of the intensity ratio of each of the lines 2.88 and 3.03 meV compared to line 3.42 meV. This shows that all the measured points fit the straight line with the slope 0.35 ± 0.1 meV. This value is apparently characteristic of the energy difference between the two levels.

As can be seen from Fig. 2, an abrupt decrease of the intensity all over the spectrum occurs at temperatures less than 2.5 K. The investigative results of this phenomenon will be the subject of the last section of this report.

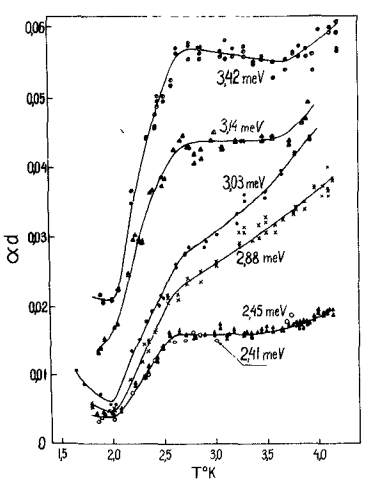

Fig. 2. The temperature dependence of the absorption intensity in different parts of the exciton spectrum (excitation level 360 mW).

III. ENERGY SPECTRUM OF INDIRECT EXCITONS IN GERMANIUM

Since the structure of exciton energy levels due to valence and conduction bands is complex in germanium, it is a very hard problem to determine the binding energy and the whole energy spectrum of the indirect exciton from the measured absorption spectra themselves. As in the case of shallow impurities the most precise results can be obtained by comparing the experimental data with the results of theoretical calculations, especially for the excited states [9]. The binding energy values estimated directly from the interband transition experiments [10], [11], or using the simplest hydrogen-like model and ignoring the energy-band complexity of germanium [12], are rather inaccurate. For the present time the most precise calculations by variational methods have been carried only for the two lowest 1s states of indirect exciton in germanium at 2.9 and 3.47 meV [10].

In order to determine the value of exciton binding energy more precisely and to estimate the whole energy structure of indirect excitons in germanium from the measured absorption spectra we use the approximation [11], which makes it possible to reduce the exciton problem to the two different and more simple shallow donor-like problem by neglecting the nondiagonal members in the effective-mass exciton equations. Each of these two problems can be solved separately by means of the variational method. The energy values of the two lowest exciton states in germanium, estimated by the use of this method, 2.5 and 3.3 meV [11] are in good agreement with the results of more accurate calculations [10]. In Table I we present the results of energy-level calculations obtained by this method for the two exciton problems, namely, for "isotropic" and "anisotropic" excitons, differing by the value of anisotropy factor $\beta = (m_T/m_L)^{1/3}$. Instead of the direct variational calculation we used an interpolation procedure, based on the known variational solutions for the shallow donors in germanium ($\beta = 0.37$) [13] and

TABLE I
ENERGY LEVELS OF INDIRECT EXCITON IN GERMANIUM

"Isotropic" exciton $\beta = 1.085$				"Anisotropic" exciton $\beta = 0.445$					
$1s$	$2s, 2p$	$3s, 3p$		$1s$	$2p\,0$	$2s$	$3p\,0$	$2p\,\pm 1$	$3p\,\pm 1$
Energy, meV	2.883	0.721	0.317	3.711	1.654	1.250	0.846	0.673	0.34

Note: Estimated in approximation [11] by the use of an interpolation procedure and parameters [17]–[19].

for the shallow donors in silicon ($\beta = 0.575$) [14]. If, as in the case of shallow-donor impurities in germanium, the most intense exciton absorption lines correspond to $(1s) \rightarrow (2p \pm 1)$ and $(1s) \rightarrow (3p \pm 1)$ types of the exciton transitions, it is rather natural to attribute the most pronounced absorption lines at 3.14 and 3.42 meV to these transitions. The estimated exciton binding energy at these assumptions is about 3.8 meV in agreement with the photoconductivity data [4], [15] and the main exciton transitions predicted by the model are in the spectral region of the measured exciton absorption.

According to the interband transition experiments [10]–[12], as well as theoretical calculations [10], [11], including the approximate model used in the present work, the ground-state splitting for the indirect exciton in germanium is estimated to be ~0.8 meV. Since the exciton transitions from the upper of these two states may be hardly observable at the temperature conditions of the experiment (1.6–4.2 K), the observed activation energy $\Delta = 0.35$ meV can be considered as corresponding to the additional splitting of the lowest ground state, possibly due to the exchange interaction [16].

We should remember that our estimated binding energy and energy-level structures of the indirect exciton in germanium are rather approximate. The more precise information about the energy structure of excitons in germanium can be obtained by comparing the experimental data with the results of more accurate theoretical calculations, but the real exciton binding energy seems to be not less than the value estimated.

IV. PHASE TRANSITION OF EXCITONS INTO THE CONDENSED STATE

As was pointed out, under temperatures lower than 2.5 K we observed an abrupt decrease of the intensity of exciton absorption all over the spectrum (Fig. 2). The measurements by the use of a diffraction spectrometer show that an intense resonance-type absorption with maximum at ~8.9 meV appears approximately under the same temperatures. As was established earlier [20], this absorption is due to the formation in the excitonic system of a condensed phase, namely metal-like electron-hole drops [20]. The observed changing of the intensity of exciton absorption under low temperatures is obviously a consequence of exciton condensation into this new phase. Measurements under two different excitation levels confirm the threshold character of the exciton absorption dependence on temperature (Fig. 4). One can see that the threshold temperature becomes lower as the excitation level decreases, which should be expected in the case of phase transition. In Fig. 5 we present the comparative measured data on the temperature dependence of the resonance absorption intensity proportional to the whole number of carriers in the condensed state and the exciton absorption intensity, taking into consideration the relative population of the two lowest states of excitons. This comparison evidently shows the common point for the both curves, temperature threshold beyond which the system exists as an excitonic gas and under which the main portion of carriers passed into the condensed state. Using these temperature dependencies $(\alpha d)_c$ and $(\alpha d)_{ex}$ it is possible to estimate the value of the work function for transition of carriers from the condensed state into excitonic gas $\varphi \simeq 1.0$ and 1.5 meV, which could be considered as quite reasonable [21].

The results presented here of the submillimeter absorption study also prove that the exciton phase transition into the condensed state takes place without the formation

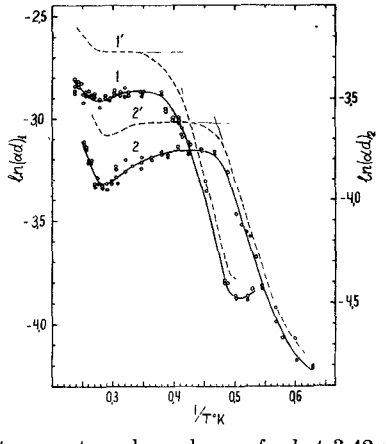

Fig. 4. The temperature dependence of αd at 3.42 meV under two different excitation levels: 1—360 mW and 2—140 mW. The dotted lines show the calculated numbers of free excitons N_{ex} taking into consideration the relative population of the two lowest exciton states.

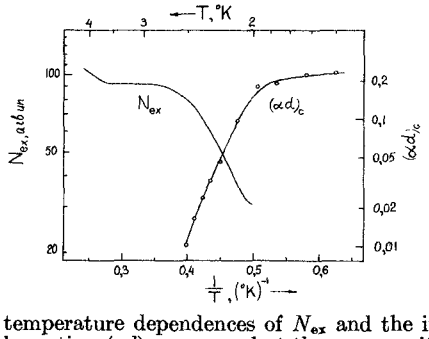

Fig. 5. The temperature dependences of N_{ex} and the intensity of resonance absorption $(\alpha d)_c$, measured at the same excitation level 360 mW.

of weak-bounded excitonic molecules. Otherwise an additional absorption structure due to these complexes would appear in the exciton absorption spectrum as the temperature decreases.

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Waveguide and Open-Resonator Techniques for Submillimeter Waves

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***Abstract*—Resonance techniques at 337 μm (890 GHz) are used to study the performance of modified *H* guide as a low-loss waveguide and of open resonators using spherical mirrors for media characteristic measurement.**

I. INTRODUCTION

THE exploitation of the submillimeter wave region will require the development of a low-mode waveguide of relatively low loss. Consideration of the various guide possibilities has led to *H* guide as the most likely structure [1], [2]. This guide uses the low-loss mode between parallel conducting planes, with a thin dielectric film to concentrate the propagated energy in the required direction. Modifications are required to give a design which combines feasible construction and low loss. Previous measurements of submillimeter waveguide attenuation are confined to loss determination on grossly overmoded rectangular waveguides at 0.85- and 0.65-mm wavelengths. The characteristics of the guide described in this paper have been investigated at 3 cm, 8 mm, and 0.34 mm using a resonance technique, the latter measurements using a HCN laser as source.

Resonance spectra also form the basis for measurement of propagation characteristics of media at 890 GHz by open-resonator techniques. Such methods have been used

Manuscript received May 6, 1974. This work was supported by the United Kingdom Science Research Council.

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