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Optical Properties of Extrinsic Two-Dimensional Excitons in BiI_3 Single Crystals

Katsuyoshi WATANABE,* Tsutomu KARASAWA, Teruo KOMATSU
and Yozo KAIFU

Department of Physics, Osaka City University,
Sumiyoshi-ku, Osaka 558

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Interface excitons caused by stacking faults in BiI_3 crystals have been investigated on absorption, photoluminescence and its excitation spectra. Characteristic three sharp absorption lines (called R, S and T), appear close to the fundamental absorption edge. The temperature dependence of the line shape shows a typical feature of exciton-phonon interaction in the case that $K=0$ at the bottom of the exciton band. Resonant luminescences of these lines have also very narrow line widths, and no Stokes shifts can be detected. The luminescence line shape of phonon side band shows Maxwellian-like having the line width proportional to thermal energy but accompanied by a fine structure which reflects a quasi two-dimensional band nature of this exciton system. The high density excitation effects of these states reveal a specific interaction among the R, S and T excitons.

§1. Introduction

The electronic states occuring near planar interfaces in crystals have long been the subject of theoretical interest; for general reviews see Jones[1] and Cohen.[2] These include surfaces, semiconductor-semiconductor interfaces, metal-semiconductor interfaces and planar defects produced by stacking faults, etc. There have been comparatively fewer studies of defect states by stacking faults. These correspond to the error of stacking sequence from the perfect crystal structure. The stacking faults should break the translational symmetry of the crystalline potential along a specific direction. In tetrahedrally bonded semiconductor, the localized interface states have been discussed theoretically by a change in structural symmetry, such as zincblende-wurtzite interfaces.[3] However, clear experimental evidence of the interface states concerning the stacking faults have not been illustrated.

Layered crystals often exhibit an amount of stacking faults which were introduced during crystal growth. Such stacking faults in layered crystals are considered to provide a nice system to investigate the electronic states of planar interfaces. In layered GaSe crystals, in the presence of stacking disorder the excitons are confined to a finite number of layers, resulting in a observation of pronounced fine structure near the exciton ground state energy on the absorption spectra.[4-7] In BiI_3 crystals, we have showed that the sharp absorption lines near the absorption edge are due to excitonic transitions perturbed by some stacking faults.[8,9]

BiI_3 belongs to a family of layered metal halides. These materials occur as strongly bonded two-dimensional X-M-X layers with weak interlayer coupling by the van der Waals force. In BiI_3 crystals, the metal ions (M) occupy one-third of the octahedral holes formed by adjacent halogen (X) planes, and the M-plane consists of a honeycomb-like arrangement of Bi ions. There are three different ways of the octahedral-hole occupation constructing three different honeycomb Bi planes. In the normal stacking, the three different layers are successively stacked along the crystal c axis leading to the conventional hexagonal lattice, although the primitive cell is the rohmbohedron with space symmetry group C_{3i}^2. The optical absorption edge of BiI_3 con-

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* Present address: Department of Education and Liberal Arts, Yamanashi University, Takeda, Koufu City 400.

sists of indirect exciton transition assisted by three different momentum conserving phonons, called A, B and C.[8] The indirect exciton transition (the energy $E_{gx}^{i}=2.0081$ eV at 2 K) occurs very close to the direct exciton energy ($E_{gx}^{d}=2.072$ eV), and shows an extraordinary large oscillator strength; about 10^3 times larger than that in usual indirect semiconductors. This specific nature of large transition probability may emphasize various optical processes associated with the indirect absorption edge in BiI_3.

Sharp absorption lines in BiI_3 are often observed just below the indirect edge, and are called P, Q, R, S and T lines from high energy side. At first, in view of the fact that the P, R, S and T lines fit in well with an inverse hydrogen like series (IHS)*, Gross and his coworkers had explained these lines as due to a formation of bi-electron (or bi-hole), i.e. a two-particle system having a negative reduced mass.[10–12] However, since the occurence and the intensities of these lines depend on samples, they have been considered to be due to some extrinsic origin, and difficulties in the bi-electron model have been pointed out by several authors.[13–16]

The origin of the P line is different from that of the R, S and T lines, since the P line appears independent of the other lines. The energy position of P (2.0080 eV) is very close to the indirect exciton energy E_{gx}^{i}. In a previous paper,[17] we showed that polytypic stacking disorders fold back an exciton energy band as well as phonon dispersion curves in the Brillouin zone. Thus the indirect exciton state is folded back to the Γ point giving a new direct exciton transition in a BiI_3 crystal. The P line just corresponds to such transition.

The absorption coefficients of the R, S and T lines vary from sample to sample, but the intensity ratios among them are nearly constant and are found to be about 4: 2: 1. A series of absorption measurements following successive cleavages on several crystals show that the defect responsible for these absorption lines are not uniformly distributed along the crystal c axis. On the other hand, the absorption coefficients are nearly constant for measurements at various points on the crystal surface perpendicular to the c axis.[9] These results show that the defect is a plane-like one perpendicular to the c axis. According to crystallographic measurements by X ray, crystals showing the R, S and T lines give extra diffraction spots corresponding to a reverse sequence of the three-layere stacking. Thus some kind of stacking faults should be occur at the boundary between the normal and the reverse stacking in such a crystal. The R, S and T lines appearing near the absorption edge are considered due to excitonic transitions perturbed by such stacking faults.[8] In a recent paper,[18] we have reported that these excitons show high density effects under much lower excitation intensity compared with the case of the intrinsic bulk exciton, because they are confined into a quasi two-dimensional space at the stacking faults, and interact each other.

In this paper, we report detailed experimental results of the absorption and luminescence spectra of the R, S and T lines, and discuss the specific properties of these two-dimensionally trapped excitons at the stacking faults in BiI_3.

§2. Experimental Procedures

BiI_3 was synthesized by reaction of iodine with elemental bismuth in a sealed tube, and was purified by sublimation in vacuum several times. The single crystal was grown by sublimation method under conditions of excess iodine in the gas phase.

In optical measurements, thin plate-like crystals with very flat surface were selected from as grown single crystals of good quality. The major crystal face is (001), and the optical measurements were made along the c axis of the crystal. The samples were immersed in liquid or gaseous helium bath, or mounted on a cold finger of a conduction-type cryostat. Thus the optical measurements were carried out from 2 K to 40 K. The sample temperature was measured by a calibrated carbon resistor and an AuFe: Chromel thermocouple placed in the vicinity of the sample.

Luminescence spectra were excited by a 45 mW He-Cd laser (4416 A line). For the measurements of high density effects, a dye

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* The P, R, S and T lines had been attributed to the $n=3$, 4, 5 and 6 lines, respectively, in the IHS.

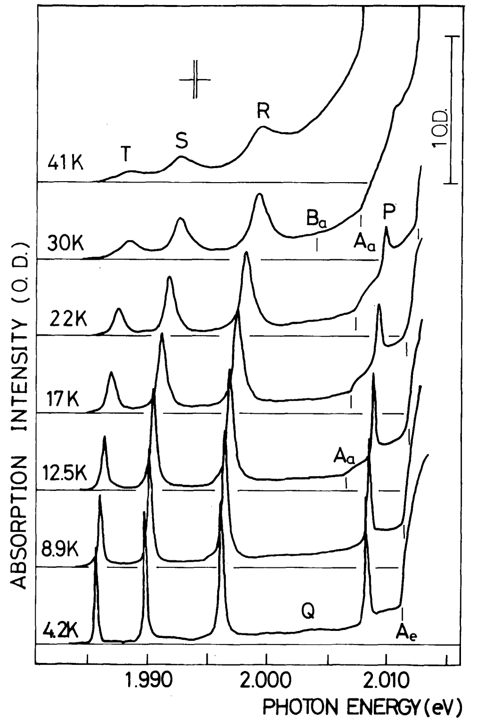

Fig. 1. Absorption spectra of BiI_3 single crystal at various temperatures. *R*, *S* and *T* are the extrinsic two-dimensional excitons. A_e, A_a and B_a are the threshold energies of indirect exciton transitions assisted by *A* and *B* phonon emission (sufix *e*) and absorption (*a*). *P*: the exciton transition due to polytypic structure; the origin of *Q* is not clear.

laser system pumped by a pulsed nitrogen laser (MOLECTRON Model UV-12: DL-12) was used. Absorption and luminescence spectra were analyzed with a 75 cm single grating monochromator (SPEX Model 1704) and a 60 cm double grating monochromator (JOBIN YVON Model HRD-1). The resolution of the analyzing system in the present work was up to 0.25 A.

§3. Experimental Results

Figure 1 shows absorption spectra near the indirect absorption edge for different temperatures. At 4.2 K, sharp absorption lines *P*, *R*, *S* and *T*, and a small hump *Q* can be seen in the low energy side of the intrinsic absorption step A_e which corresponds to the indirect exciton transition assisted by the smallest A_g-mode phonon (called *A* phonon) emission. The intensities of the *R*, *S* and *T* lines depend on samples, i.e. the amount of stacking faults in the samples. The absorption coefficients reach more than 1.5×10^3 cm^{-1} in usual thin crystals. Previously in extremely thin crystals, we showed that the estimated oscillator strengths of these lines are comparable to that of the direct exciton transition.[19] The peak wavelengths of *P*, *Q*, *R*, *S* and *T* lines at 4.2 K are found at 6172.7, 6188.2, 6209.6, 6229.2 and 6242.2 A, respectively. Many authors have observed a broader line at 6418 A in absorption as well as emission spectra at liquid helium temperature (LHeT).[16,20,21] In a previous paper,[9] we showed that the 6418 A line can be ascribed to a center created by effects of strains in the crystal. Occasionally, samples have extra line at 6255 A, not appeared in Fig. 1, due to unknown origin. In the present investigation, we selected the crystals having the *R*, *S* and *T* lines.

The line shapes of *R*, *S* and *T* lines are very sharp and appear to be symmetric below 4.2 K being restricted by the resolution of the apparatus. The line widths of *R*, *S* and *T* in a typical sample reach to be 0.4, 0.3 and 0.2 meV, respectively. But in some sample, the widths are much broader, depending on the sample quality. As the temperature increases, the line shapes become broad and asymmetric having the tails at higher energy side, and besides, the intrinsic transition of indirect exciton assisted by phonon absorption appears as a background component. The A_a and B_a in Fig. 1 denote the threshold energies of the indirect exciton transition accompanied by the *A* and *B* phonon absorptions. Above 50 K, the intrinsic absorption part blures out the sharp absorption lines and approaches to so called Urbach tail.[8] All these thresholds and the sharp lines shift toward higher energy side with increasing temperature. However, the temperature coefficients of the sharp lines are slightly larger than that of the indirect exciton gap.[8]

The temperature dependences of the half value widths of *R*, *S* and *T* lines are summerized in Fig. 2. Each width is nearly constant below 5 K, and increases linearly with increasing temperature between 6 and 25 K. The widths at lowest temperature depend on samples, and in some crystals, they show several times larger values. However, the temperature

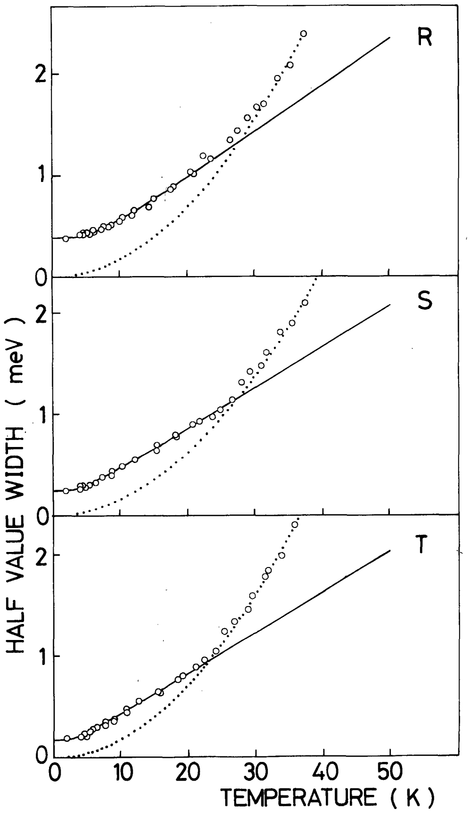

Fig. 2. Temperature dependences of the half value widths of absorption lines R, S and T. The solid lines are the best fitting curves of eq. (1) with $H_0=0.39$, 0.25 and 0.17 meV and $\hbar\omega=1.44$, 1.03 and 0.72 meV for R, S and T, respectively. The dotted lines are those of the square of the temperature with coefficient values of 0.0017, 0.0015 and 0.0018 meVK^{-2} for R, S and T, respectively.

dependences of width-changes are common. Above 25 K, the widths increase much more away from the linear relation and become to fit the square of the absolute temperature as shown by dotted curves in the figure.

Figure 3 shows a typical luminescence spectrum at 4.2 K around the sharp absorption lines under the excitation with band-to-band light. The spectrum consist of the indirect exciton recombination band L_C assisted by C phonon, the resonant luminescence lines R, S and T, and phonon side bands T_B, S_C and T_C of S and T luminescences, where the suffixes denote the phonons concerned. Additional luminescence lines are observed in the lower energy side of the resonant line T. These depend drastically on samples, and one of them has been associated to the extra absorption line at 6255 A.[20] But these are not discussed in this paper.

The line shapes of resonant luminescences R, S and T are very sharp, and their widths are less than 0.5 meV at 4.2 K. The peak energies of these lines are equal to those of the respective absorption lines. Thus the Stokes shift can not be resolved within the experimental resolution. The intensities of resonant luminescence lines are large in order of T, S and R, contrary to the absorption coefficients, and the ratios are typically observed to be about 100: 10: 1 at 4.2 K. This result is interpreted as mainly due to a cascade type transfer of the excitation energy among them, which can be assured by excitation spectra shown in Fig. 4.

The excitation spectra for the luminescences T and T_C at 4.2 K give nearly the same order response intensities peaked exactly at the S, R and Q, and the T, S, R and Q absorption lines, respectively. The resolution is not enough but the R, S and T response spectra clearly show an asymmetric line shape having a tail at higher energy side. In Fig. 4, the P line appears as a response peak, but in some samples, appears as a dip. The insert shows the excitation spectrum for T in wide energy range. This shows response minimum at the direct exciton energy E_{gx}^{d}. Such feature has been usually observed in excitation spectra related to direct-exciton decays in many other crystals. On the other hand, the indirect exciton absorption edge does not show clear response in the excitation spectrum for the R, S and T luminescences.

Figure 5 shows the temperature dependences of the luminescence spectra of R, S and T lines. As the temperature increases, each line becomes broad, and the line shape is asymmetrically tailed to the lower energy side. Each luminescence intensity decreases with increasing temperature, and the reduction rates are large in order of T, S and R. Thus the intensity differences among them become small at higher temperature. Each peak energy shifts to higher energy side in the same manner as in the absorption and the Stokes shift also can not be observed. The tail parts of both absorption and luminescence lines become mirror symmetric to each other at higher

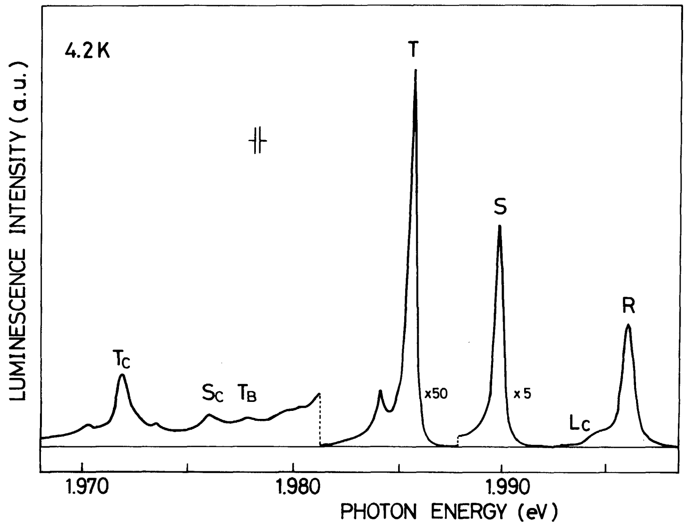

Fig. 3. Luminescence spectrum of BiI_3 single crystal excited with a He-Cd laser (4416A line) at 4.2 K. The resonant luminescence lines R, S and T appear. L_C: recombination luminescence band of indirect exciton assisted by the C phonon; S_C, T_B and T_C: phonon side bands of luminescences S and T assisted by the B and C phonons.

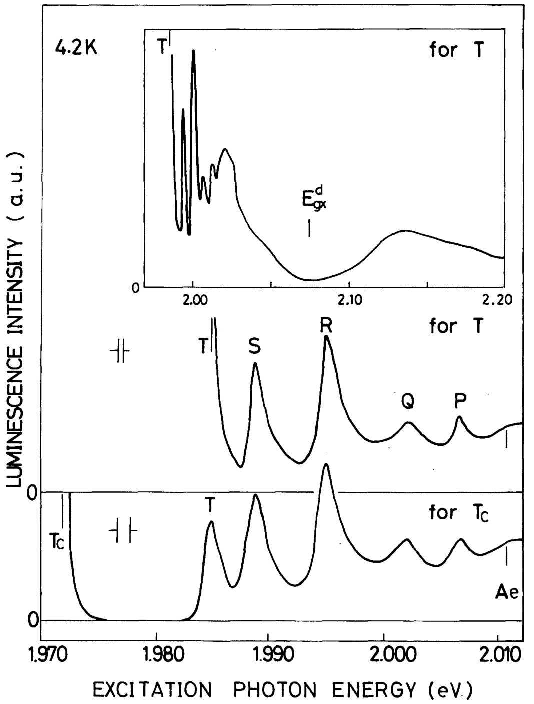

Fig. 4. Excitation spectra for resonant luminescence line T and phonon side band T_C at 4.2 K. The insert shows the same excitation spectrum for T with wider energy range; E^d_{gx} the direct exciton energy.

temperature.

A line shape of phonon side band of exciton luminescence reflects the exciton distribution by the center-of-mass motion in the band. We carefully investigated the shape of T_C band, and the results are shown in Fig. 6. The line shape is broader than the resonant luminescence line T, and shows the Maxwellian-like shape having the width proportional to the lattice energy kT. These results were already pointed out by the Leningrad group in connection with the bi-electron model.[20] In more detail, the half value widths at 2.1, 4.2, 5.0 and 10 K are 0.51, 0.68, 0.89 and 1.97 meV, respectively. By the careful measurement, a small hump can be resolved at higher energy side by about 0.4 meV from the main peak below 4.2 K. The hump appears independent of samples, and is considered to be an intrinsic structure of the T_C line. The energy separation is in agreement with the energy dispersion of C phonon branch between the Γ and the Z points in the Brillouin zone, which has been observed to be 0.39 meV in the resonant Raman scattering measurements.[22]

Finally we present an additional result on the high density excitation effects of the R, S and T states. As previously reported, under the resonant excitation at these absorption lines by a high power laser light, the peaks of these lines shift toward higher energy side, and the line shape becomes broad and asymmetric

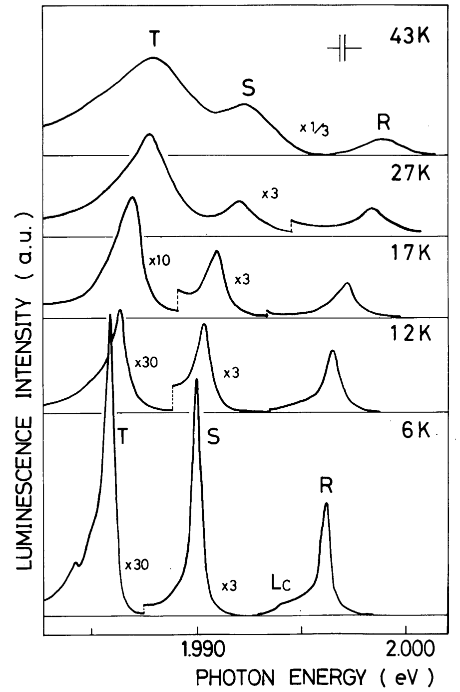

Fig. 5. Luminescence spectra of R, S and T lines at various temperatures excited with the 4416A line.

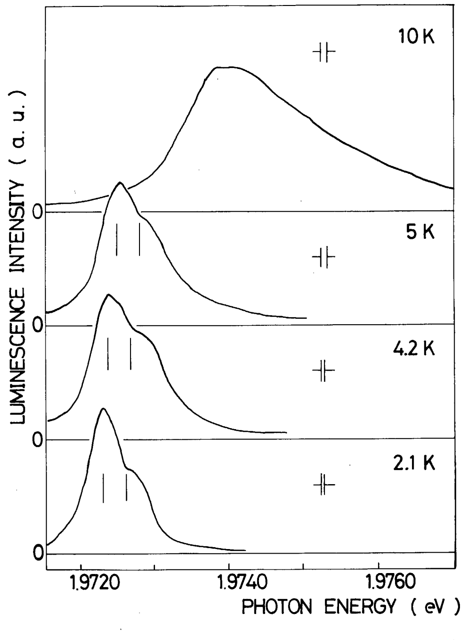

Fig. 6. Luminescence spectra of phonon side band T_C in BiI_3 at different four temperatures. The vertical bars denote the energy separation of 0.39 meV corresponding to the C phonon dispersion at the Γ and the Z points (see text).

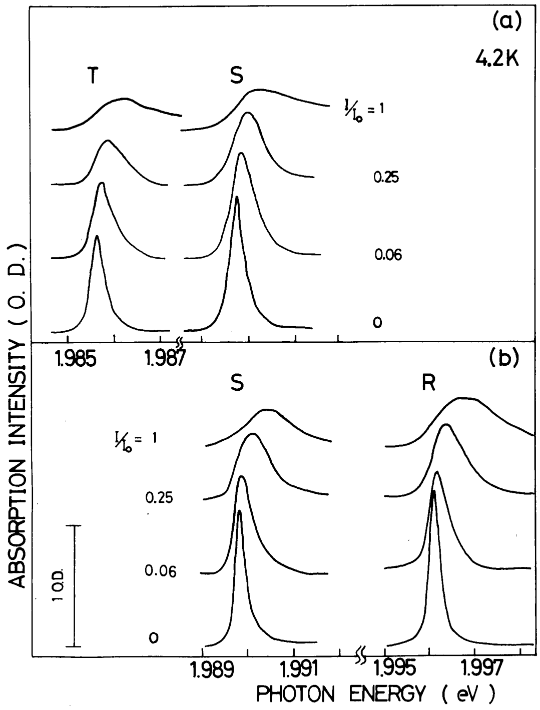

Fig. 7. Absorption spectra of R, S and T lines under the high power laser excitation of various intensities (a) tuned to the highest lying R state and (b) to the lowest lying T state. I_0: the maximum laser intensity; I: respective reduced laser intensities (see text and ref. 18).

spreading to higher energy side with increasing excitation density.[18] Figure 7 shows the absorption spectra of R, S and T by the two-beam (excitation and probe) transmission measurements under two different excitation frequencies tuned to (a) upper lying R state and (b) lower lying T state. Details of the two-beam method have been described in the previous report.

As the excitation density (I/I_0) increases, all the absorption lines show the peak shift and the asymmetric broadening as seen in Fig. 7. The peak energy shift $\Delta\Omega(=\Omega-\Omega_0)$ and the line broadening $\Delta\Gamma(=\Gamma-\Gamma_0)$ are plotted as a function of excitation density in Fig. 8 under the R excitation (a) and the T excitation (b). Where, Ω and Γ show peak energy and line width under the laser excitation; Ω_0 and Γ_0 those values obtained from conventional absorption measurements. The excitation density is corrected for the change of absorption intensities of R and T lines under the high densi-

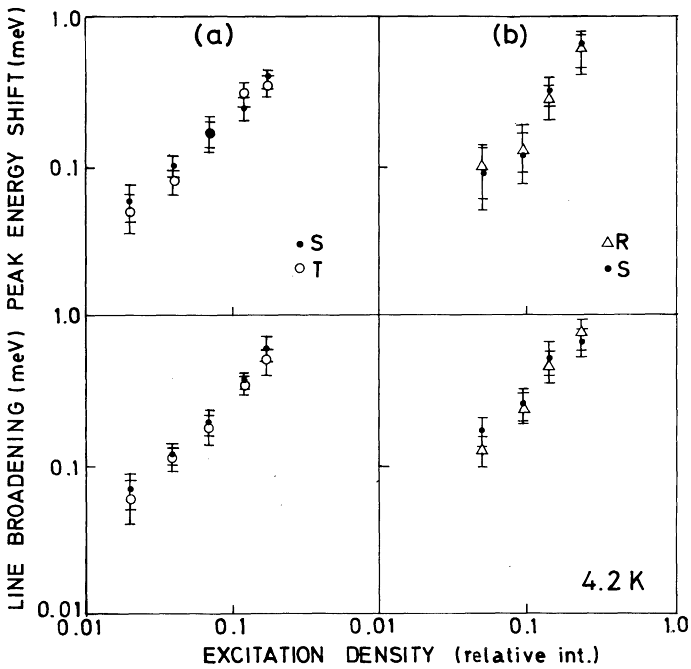

Fig. 8. Peak energy shifts and line broadenings of *R*, *S* and *T* lines under the *R* excitation (a) and the *T* excitation (b) at 4.2 K as a function of relative intensity of exciting laser light.

ty excitation. The results under the *R* excitation have been already reported. The same nearly linear dependences of $\Delta\Omega$ and $\Delta\Gamma$ for excitation density can be seen even under the *T* excitation. It should be noted that the high density effects in *R*, *S* and *T* states can be observed equally under the resonant excitation of any one of these states. Under the excitation of *T*, the excitons in *R* and *S* states can not be created because the excitation photon energy is too short. Thus the phenomena cannot be explained as due to a simple exciton-exciton collision in the same state under high density. We have to consider a renormalization of exciton self-energy caused by the existence of *R*, *S* or *T* excitons at the stacking fault layer. The importance of the dielectric exciton screening effect in exciton-exciton interacting system were pointed out in recent observations in bulk GaAs[23)] and in multiple quantum well structure in GaAs.[24)]

§4. Discussions

The *R*, *S* and *T* states are considered to be exciton-polariton states excited around a stacking fault in BiI_3 crystals. The diameter of the ground state exciton in the host crystal has been estimated to be comparable to the thickness of I-Bi-I unit layer.[25)] As the stacking fault effects spread over a few unit layers, it is expected that the *R*, *S* and *T* excitons propagate freely along the layer of the stacking fault and the size quantization may not be important. However, they are surely localized along the axis of non-periodicity. The result that such localizations of excitons are produced by stacking disorder itself has been discussed related to fine structures of exciton absorption band observed in GaSe.[6)]

The two-dimensional natures of *R*, *S* and *T* states have been revealed in our previous paper,[18)] and some more clearer evidence can be seen in the line shape of phonon side band T_C shown in Fig. 6. The line shape shows Maxwellian-like having a width proportional to thermal energy *kT*. This result shows that the *T* exciton is in center-of-mass motion and is in thermal equilibrium with lattice. However, the hump resolved in the high resolution spectra can not be explained from the Maxwell-Boltzmann distribution. We can give a following interpretation based on the two-dimen-

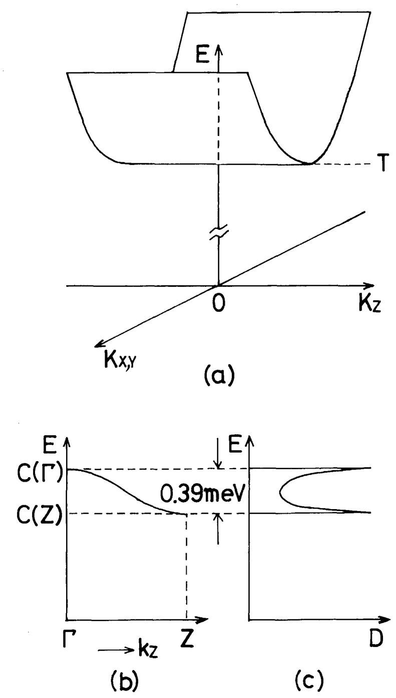

Fig. 9. (a): Schematic representation of the energy band of the extrinsic two-dimensional exciton state T. (b) and (c): schematic representation of the dispersion curve of C phonon along the $\Gamma-Z$ line and corresponding density of state D.

sional exciton band. If the two-dimensional exciton is a limit of anisotropic exciton with an infinite mass in the K_z direction, the T state can be regarded as such excitons having a parabolic band in the direction of K_x and K_y, but the energy is constant in the K_z direction as shown in Fig. 9(a). In the radiative recombination process of excitons assisted by phonons, momentum selection rule for the K_z direction is no longer required, and all of the C-phonon branch with any K vectors along the $\Gamma-Z$ line can contribute to the process. According to the result of resonant Raman scattering,[22] the dispersion and the density of state of C phonons along the $\Gamma-Z$ direction can be assumed as shown in Fig. 9(b). The density of state of C phonons has two peaks with energy separation of 0.39 meV. Thus the fine structure observed in the phonon side band T_C can be interpreted in connection with the quasi two-dimensional band of T exciton shown in Fig. 9.

Apart from the two-dimensional nature, the excitons of R, S and T states give advantageous systems to study free exciton-polariton behaviors. Owing to the extremely large absorption coefficient, optical investigations of direct transition of bulk excitons in usual semiconductors encounter various difficulties related to surface, imperfection and so on. Recently, double heterostructure such an AlGaAs-GaAs-AlGaAs have been used to overcome these difficulties. Even in these systems, however, interface defects play as dominant exciton trapping centers, and Stokes shift of a few meV has been found between the first exciton absorption and the luminescence peak.[26,27] Furthermore, both absorption and luminescence lines show spectral width of several meV at LHeT, which is ascribed to an inhomogeneous broadening.

On the other hand, the R, S and T lines have very narrow line widths (about one tenth as small in the above case) with adequate absorption strength and strong luminescence intensity, and Stokes shift between the absorption and the resonant luminescence lines cannot be detected within the experimental resolution. Thus the stacking fault in BiI_3 crystals constructs an ideal plane space, which allows us to study the optical spectra of free exciton-polariton behaviors. This situation is due to the fact that stacking faults do not involve the breaking of the nearest neighbor bonds. Therefore, one can expect that the energies, wavefunctions and charge distributions of such stacking fault states will be only mildly perturbed from those in a perfect crystal. Furthermore, in layered crystals, the situation is more favorable because the interlayer bonding is weak van der Waals forces different from the interface of heterojunctions characterized by the interfacial chemical bonds of covalent and ionic natures. From these specific natures of the present exciton-polariton system, we discuss the experimental results on R, S and T spectra.

The sharp absorption lines at LHeT become broad and asymmetric tailing toward higher energy side with increasing temperature. The temperature dependences of the half value widths below 25 K can be apparently fitted with following equation as done on exciton spectra in alkali halide crystals;[28]

$$H = H_0 \coth(\hbar\omega/2kT), \tag{1}$$

where H_0 denotes the width at 0 K, ω the effective phonon frequency. The effective phonon energies are obtained to be about 1 meV and are very small as less than the smallest optical phonon energy of 1.6 meV.[22] It is suggested that the phonons dominantly interacting with these excitons are acoustic-mode ones. However, as mentioned before, the values of H_0 must be changed very much depending on samples. Then the line widths at the lowest temperature are considered to be governed by the inhomogeneous broadening.

Above about 25 K as shown by the dotted curves in Fig. 2, the half value widths are rather proportional to the square of the absolute temperature. According to the theory developed by Toyozawa,[29] in case of the lowest exciton band where the $K=0$ state is at the band bottom and where there exists no levels of the same energy in other bands, the contributions from higher order scatterings by acoustic phonon are predominant and the line width is proportional to the square of the absolute temperature within a certain temperature range. The situation is favorable in the present exciton system, because the T exciton is created by photons in an extremely narrow energy range at $K=0$ restricted by an inhomogeneous broadening. The asymmetric line shape at high temperatures is also interpreted along this line. We observed, for the first time, similar T^2 dependence of exciton absorption spectra in the lowest exciton state in CuCl crystals.[30] However, more experimental and theoretical investigations in the present system are needed for further insight of the line width as well as the line shape at whole temperature range.

The line shape of resonant luminescence also changes drastically with increasing temperature. The width at LHeT is very sharp as in the absorption spectrum. The three luminescence spectra are compared in Fig. 10; the indirect exciton recombination luminescence band L_C assisted by the C phonon, the resonant luminescence T and the phonon side band T_C of the T line assisted by the C phonon. The L_C is Maxwellian-like, but the line width is several times larger than kT and depends on the excitation energy. These hot luminescence natures of L_C were discussed in previous papers.[31,32] As mentioned before, the width of T_C is nearly kT showing that the T exciton is in thermal equilibrium with lattice before the radiative recombination. On the other hand, the resonant luminescence shows

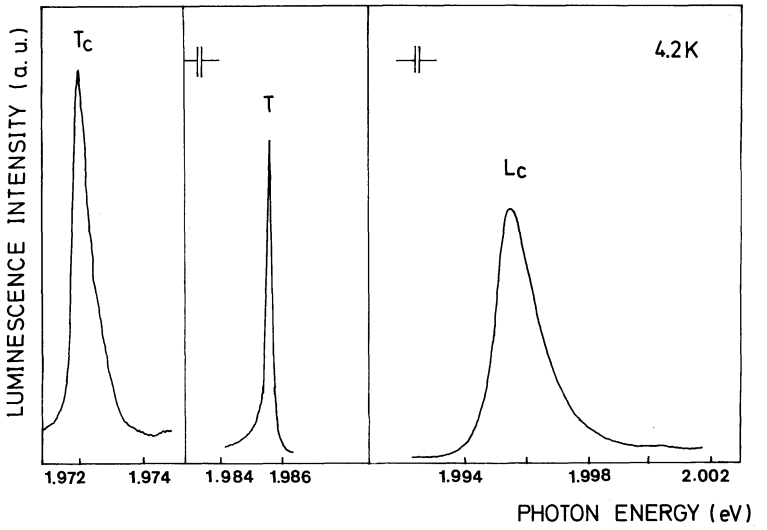

Fig. 10. Comparison of the three luminescence spectra in BiI_3 single crystal at 4.2 K; the recombination luminescence band L_C of indirect exciton assisted by the C phonon, the resonant luminescence line T and the phonon side band T_C.

extremely narrow line width at 4.2 K. The difference between T and T_C line shapes clearly shows that the K-conservation rule plays an important role in the luminescence process of direct exciton. Relaxation of the rule depends on various scattering mechanisms of non-local excitons with impurities, defects, phonons and so on. At higher temperature, phonons contribute dominantly to the temperature dependences of luminescence spectra as shown in Fig. 5. But the effects of imperfections cannot be excluded even at these temperatures, because the line shape depends delicately on samples.

The excitation spectra for T and T_C luminescences show the existence of excitation energy transfer from the higher lying R and S states to the lower ones, and the radiative recombination occurs dominantly at the T state. The transfer efficiency decreases with increasing temperature, as can be seen in Fig. 5. Thus the transfer process is not understandable to be a simple energy relaxation between upper and lowe levels, and it seems to need a consideration of some specific interactions among the R, S and T exciton states.

From the results that the intensity ratio among the three absorption lines are always constant, and that the high density excitation effects occur just parallel among them, the three exciton states are thought to be associated with the same stacking fault. Although the atomic arrangement in the unit I-Bi-I layer is not altered, the stacking-fault layers composed of a few unit layers have different symmetry of the two-dimensionality from that of the host lattice. Thus it is likely that the reduction of symmetry splits off the degenerate host exciton modes giving the three modes R, S and T. According to this speculative model, we have to consider an anharmonic interaction among the three modes confined in the two-dimensional lattice site. At present, further insight of such interaction cannot be clarified, but the results of the high density resonant excitations in Fig. 7 show that they are controlled by the density dependent polarizability, and give a renormalized self-energy of exciton to higher values. The polarizability also depends on various exciton scattering processes which decide the final process of exciton recombinations. We are now investigating the space and time dependences of dynamical exciton behaviors in this system. The details will be reported near future.

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