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Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

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Version of record first published: 04 Oct 2006.

To cite this article: T. Komatsu, T. Iiad, K. Murayama, M. Ichida, H. Kurisu, H. Kondo, I. Akai & T. Karasawa (1992): Excitons Localized in Mesoscopic Domains Produced by Stacking Fault In BiI_3 , Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 218:1, 37-42

To link to this article: <http://dx.doi.org/10.1080/10587259208047012>

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EXCITONS LOCALIZED IN MESOSCOPIC DOMAINS PRODUCED BY STACKING FAULT IN BiI_3

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Abstract Absorption line series W^J ($J=\text{I,II,III},\dots$) appeared below the indirect edge in BiI_3 has been studied. The series is assigned to the excitons confined in mesoscopic domains produced by deformation faults characteristic of layered materials. Each line consists of four structures which correspond to $x+iy$, $x-iy$, z , and pure triplet states originated from the bulk cationic excitons. The energy separations between these four states are depend on the size of mesoscopic domains in which an exciton is confined. By introducing a disk-like shape model of the mesoscopic domain, the experimental results are analyzed on the basis of anisotropic quantum size effects on the cationic excitons. The relative motion of the exciton is found to be strongly affected by the localization of the translational motion in the direction perpendicular to the confinement axis.

INTRODUCTION

Quantum size effects of the excitons have been extensively studied in semiconductors. In most of semiconductor quantum wells, like in a GaAs/AlGaAs quantum well, remarkable size effects have been known as the result of the spatial confinement of the exciton relative motion in the quasi two-dimensional space. On the contrary, in microcrystals, the confinement of the exciton translational motion occurs in a material having relatively smaller exciton radius than the size of microcrystal. In theoretical analysis of the quantum size effect of microcrystal, the shape of the microcrystals is regard as approximately spherical.^{1,2} The quantum size effect of the exciton is strongly dependent on what shape

the microcrystal has. In layered crystals having strong anisotropy of bonding nature, a characteristic quantum size effects reflecting such an anisotropic structure can be expected.

In this paper, analyzing the experimental data, we show that the excitons confined in the mesoscopic domains produced by deformation faults in layered BiI_3 crystals have a strong anisotropic size effects. The various types of the excitons in BiI_3 have been reviewed in the reference 3. Magneto-optic effects measured on the bulk⁴ and stacking fault excitons⁵ have been reported. The band edge excitons in BiI_3 are composed of four states originated from the cationic exciton states,³ the spin mixed $x - iy$, $x + iy$, z state and the pure triplet state t . In the magneto-optic spectra, the four structures have appeared due to Zeeman effect by mixing among the states under magnetic fields. The magneto-optic effects on the W-lines have been also investigated.⁶ The energy positions have been determined for the t -state of W^J and $W^{J'}$ states studied in this paper.⁷

RESULT AND DISCUSSION

Optical Spectra

The absorption spectrum below the indirect exciton edge in BiI_3 at 4.2 K is shown in Fig. 1(a) for the sample applying the bending stress before measurement. The absorption intensities of the W^J ($J=\text{I,II,III},\dots$) lines depend on the degree of applying stress. The transition energies of the W^{I} , W^{II} and W^{III} lines are observed at 1.931 eV, 1.878 eV and 1.850 eV, respectively. The W^{I} line is the same as that called the W line previously, which was observed when the crystals were created.⁸ The W^{I} line also appears by applying the shear type stress to the crystals. This suggests that the origin of the W^J transitions closely relates to the formation of the stacking disorder induced by deformation between layers. The luminescence spectrum is shown in Fig.1(b). The resonance lines are observed with no Stokes shift and the other lines $W^{J'}$ ($J'=\text{I}',\text{II}',\text{III}',\dots$) appeared on the low energy side of the resonance lines with nearly constant energy separations. The energy positions of these absorption and luminescence lines

form a series with energy separations which become narrower with going to the low energy side. This is the characteristic behavior of quantum size effects of excitons confined in microdomains with different sizes.

Figures 2(a), (b), (c) and (d) show the excitation spectra for the W^I , W^{II} , $W^{I'}$ and $W^{II'}$ lines, respectively. The excitation spectra for the W^I , W^{II} , $W^{I'}$ and $W^{II'}$ lines show response to each associated state whose orbital symmetry is z -like (indicated by arrows in Fig.2). This suggests that there exists a strong energy relaxation process among internal structures of the cationic states. The excitation response for the $W^{I'}$ line is observed at the W^I and that for the $W^{II'}$ line is observed at the W^{II} state.

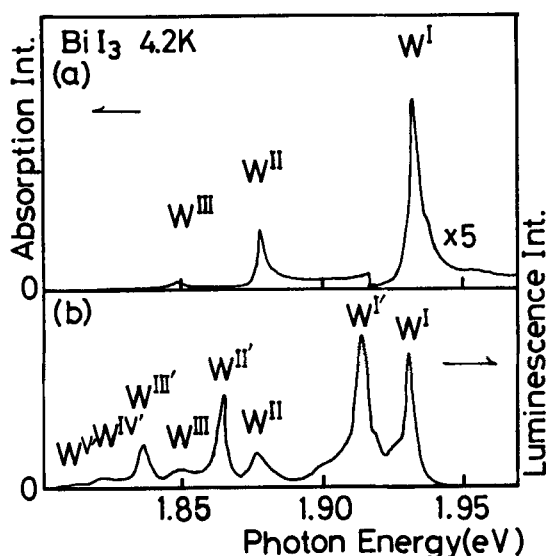


FIGURE 1 Absorption (a) and luminescence (b) spectra at 4.2K for the BiI_3 sample applying the bending stress before measurement.

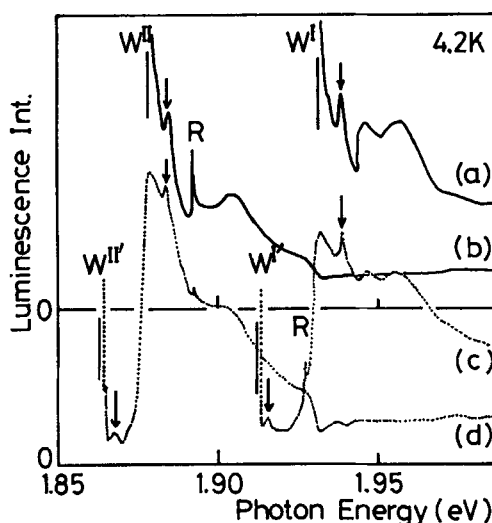


FIGURE 2 Excitation spectra for the W^I (a), W^{II} (b), $W^{I'}$ (c), and $W^{II'}$ (d) lines, respectively. R is the Raman response.

There exist energy transfer mechanisms from W^I to $W^{I'}$ and from W^{II} to $W^{II'}$ state. This suggests that the dashed states of $W^{J'}$ are located on the low energy side of the W^J states. No response of the excitation spectra have been observed for the W^{II} and $W^{II'}$ lines exciting at the W^I and $W^{I'}$ state. There is no evidence of direct energy transfer between different states localized around deformation faults distributed at the different spatial positions.

Deformation Fault

In layered crystals, various types of the stacking faults are often formed in the process of the crystalline growth or by applying deformation due to weak inter-layer bonding of the layered materials. In BiI_3 crystals, the regular structure is C_{3i}^2 symmetry in which arrangement can be denoted as $\alpha\beta^A\gamma\alpha\beta^B\gamma\alpha\gamma\beta^C\gamma\alpha\beta^A\gamma\alpha\beta^B\gamma\alpha\beta^C\gamma\dots$, with the sandwiched I-Bi-I unit layer stacking sequence, where α denoted the I^- ion site, β the metal site, γ the I^- site in the cubic close packing notations. A , B and C are the name of the different metal site denoted the vacant occupation of the metal in the honeycomb lattice in three layer structure. From the X-ray structural analysis, a possibility of the D_{3d} symmetry has been suggested.⁹ When the crystals are deformed by creaving, bending, or shearing, the stacking fault may be formed by slipping among layers. The layer arrangement can be denoted as $\alpha\beta^A\gamma\alpha\beta^B\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^A\gamma\dots$, which is denoted by I, $\alpha\beta^A\gamma\alpha\beta^B\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^A\gamma\alpha\beta^B\gamma\dots$, by II, $\alpha\beta^A\gamma\alpha\beta^B\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^C\gamma\alpha\beta^A\gamma\alpha\beta^B\gamma\dots$, by III and etc. The underlined parts contain one, two and three stacking faults, and are D_{3d} symmetry. The translational motion of the excitons along the layer stacking direction is strongly affected by the nonperiodic potential due to stacking fault. It may be reasonable that the W^I , W^{II} and W^{III} transitions are correspond to those of the excitons localized around deformation faults of I, II and III,...

Cationic Excitons Confined in Mesoscopic Domains

The energy separations between $x - iy$ and triplet states Δ_t , and $x + iy$ and z states Δ_z for the W^I , W^{II} , $W^{I'}$ and $W^{II'}$ are listed in Table I with other related quantities. The magnitude of the energy separations are determined from spectral analyses of the observed structures in the luminescence spectra under

	$\Delta_t(\text{meV})$	$\Delta_z(\text{meV})$	$K_x(\text{meV})$	$J_z(\text{meV})$	J_z/J_x
W^I	8.6	7.7	100	46	4.6
W^{II}	5,6	6.8	60	32	5.4
$W^{I'}$	17.0	2,2	210	59	2.8
$W^{II'}$	14.4	1,3	180	50	2.8

TABLE I Observed energy separations. Δ_t is the separation between $x - iy$ and pure triplet states, and Δ_z is between $x - iy$ and z states for W^I , W^{II} , $W^{I'}$, $W^{II'}$ lines. K_x , J_z and J_z/J_x are calculated Coulomb and anisotropic exchange energies and ratio based on a cationic exciton model for the respective lines.

magnetic fields⁷ and excitation spectra. The separations can be calculated on the basis of a cationic exciton model with appropriate parameters.³ We can show that Δ_z depends on the ratio of the anisotropic exchange energy J_z/J_x and Δ_t also on the magnitude of Coulomb energy K_x . These values are given in Table I.

The larger value of both separations Δ_z , Δ_t for the W^I line compared with that of the W^{II} line corresponds well to the larger J_z and K_x values of the W^I line. This fact indicates that the size of the internal motions of the confined exciton state of the W^I is smaller than that of the W^{II} in both directions of intralayer and interlayer. Moreover, the large value of the J_z/J_x suggests the large anisotropic extension of the internal motions of the W^I state. The $W^{I'}$ and $W^{II'}$ states are more localized than W^I and W^{II} states. This is confirmed by the facts that

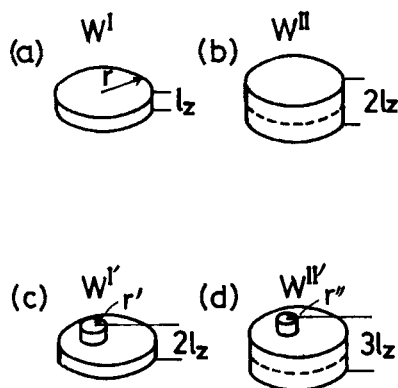


FIGURE 3 Shapes of the disk model for the transitions confined in mesoscopic domains for W^I (a), W^{II} (b), $W^{I'}$ (c), $W^{II'}$ (d).

1) the energy separations Δ_z for dashed states are smaller than that of the W^I

and W^{II} lines, 2) the estimated ratio J_z/J_x is less anisotropic than W^{I} and W^{II} lines, and 3) K_x value is large being almost twice of those for the W^{I} and W^{II} states. The small values of Δ_z in dashed states indicate the weak anisotropy of the exciton relative motions, which comes from shrinking of the relative motions by localization of the translational motions. The above experimental and calculated results are explained by the exciton states in model of microdomains as shown in Fig.(3). From above analysis, it may be concluded that the exciton relative motions are strongly affected by the translational motions in the present mesoscopic system, i.e. the stronger localization of the exciton translational motions causes shrinking of the exciton relative motions.

This work was partially supported by a Grant-in-Aid for General Scientific Research from the Ministry of Education, Science and Culture, Japan.

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