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著者	Tanabe Hisashi, Imai Isamu
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ANISOTROPY OF MAGNETO-OPTICAL REFLECTANCE OF RED-HgI₂

Hisashi Tanabe and Isamu Imai

Department of Pure and Applied Sciences, College
of General Education, University of Tokyo, Japan

Introduction

The exciton near the fundamental absorption edge of red-mercuric iodide (HgI₂) has been extensively investigated since Nikitine and co-workers¹⁾ first studied it. Nikitine et al. and Novikov et al.²⁾ observed a excitonic series (A-exciton) in absorption spectrum, and they analyzed it based on a isotropic hydrogenic model, and obtained the exciton binding energy 41 meV and 29 meV, respectively. Anedda et al.³⁾ studied the reflectivity spectrum of A-exciton with the aid of WMR method, and found structures, A₂-A₅, in the higher energy region of A₁(n=1) level. Their assignment of these structures, taking into account the anisotropy of dielectric constant and masses of exciton, was that A₂-A₄ corresponded to the 2P₀, 2S, 2P_{±1} energy levels of the exciton, and A₅ to the transition to the fourfold split n=3 level (3P₀, 3S, 3D₀, 3P_{±1}). The A₁ showed a large shift to higher energy from that expected. They explained this anomaly in terms of a repulsive central cell correction. Recently, Sakuma⁴⁾ has carried out a detailed study of excitonic structure of this material, especially of its behavior in magnetic field with the same WMR technique, and he assigned the peaks A₃, A₄, ... as LO phonon replicas of 2S exciton A₂. We present here the measurements of the anisotropic magneto-optical effect on reflectivity of red-HgI₂.

Experiment

Red-HgI₂ powder was prepared by the chemical reaction of HgCl₂ and KI. After purified by repeated sublimation in vacuum, single crystals of the size of about 8³mm³ with a smooth c-plane were grown from vapour phase in a closed growth ampulla which was placed in a furnace having temperature gradient for about one week. Fig. 1 shows the overall view of the observed reflectivity spectra of A-exciton and their changes in magnetic field. Fig. 2 shows the energies of the peaks of the reflectivity spectra as a function of the square of the magnetic field. As is seen in Fig. 2, the peaks A₂-A₅ shift proportionally to H², and are nearly parallel to each other. This suggests that the shift of the peak energies is diamagnetic and these peaks are due to the same origin. In the magnetic field of various directions with respect to the c-axis the shifts of these peaks were also observed. The results obtained in these cases were similar in nature, except that the magnitude of the shifts of these peaks

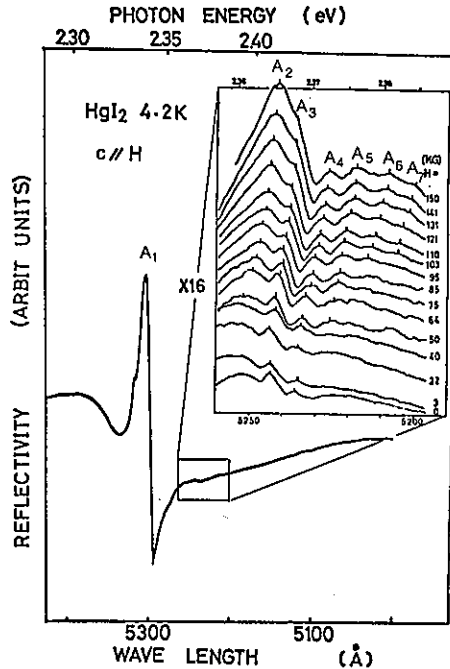


Fig 1. The reflection spectra of A-exciton of HgI_2 at 4.2K.

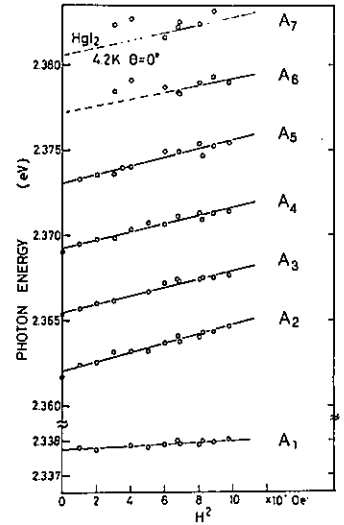


Fig 2. The energies of the peaks of the reflection of A-exciton vs the square of the magnetic field.

decreased as the c - H angle, θ , increased. This denotes that there exists anisotropy of the excitonic parameters of HgI_2 .

Discussion

According to the calculation of the excitonic energy levels,⁵⁾ when anisotropy is taken into account, the excitonic $n=2$ level splits into three levels, $2P_0$, $2S$, $2P_{\pm 1}$, and these three levels show different diamagnetic shifts, the amount of the shifts depending on the parameters c - H angle, θ , and anisotropic parameter $\gamma = \epsilon_{\perp} \mu_{\perp} / \epsilon_{\parallel} \mu_{\parallel}$. If the assignment of Anedda et al. is assumed, the ratio of the diamagnetic shifts, $\Delta E(A_2=2P_0) : \Delta E(A_3=2S) = 6:14$, and the splitting of A_4 peak are expected. Present measurement, however, showed that $\Delta E(A_2) : \Delta E(A_3) = 1:1$ and A_4 had no indication to split by application of magnetic field. These results are rather consistent with the Sakuma's assignment, $A_1(1S)$, $A_2(2S)$, $A_3(2S+1LO)$, $A_4(2S+3LO)$, \dots . According to the Wheeler and Dimmock's theory,⁶⁾ the measurement of the θ -dependence of the diamagnetic shift of the peak permitted us to make a rough estimation of $\gamma' = \mu_{\perp} / \mu_{\parallel}$. As A_3 peak is rather sharp, we had an

attempt to obtain γ' from the θ -dependence of the diamagnetic shift of A_3 peak, assuming A_3 peak is 2S like level, and got $\gamma'=0.7$. Using the observed value 2.36 meV of the diamagnetic shift of $A_3(n=2+lLO)$ at $H=100$ KOe as well as $\gamma'=0.7$ and $Ry^*=32$ meV, $\mu_{\parallel}=0.25 m_0$, $\epsilon_{\perp}\epsilon_{\parallel}=74$ and $\mu_{\perp}=0.17 m_0$ were obtained.

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