Coulomb correction to the dressed exciton in an inorganic-organic layered semiconductor: Detuning dependence of the Stark shift

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(Received 14 November 2003; revised manuscript received 2 February 2004; published 8 April 2004)

We report the detuning dependence of the excitonic optical Stark shift in the inorganic-organic layered semiconductor $(C_6H_5C_2H_4NH_3)_2PbI_4$ as a typical material in which the effective Coulomb interaction is strong. We show the polarization dependence of the *detuning spectra*, i.e., the shifts as functions of continuously changing pump photon energy, over the range of 350 meV below the exciton resonance. It is found that in the opposite-circularly and colinearly polarized configurations for pump and probe lights, the shift changes its sign at 50 meV below the exciton resonance and is resonantly enhanced around it. With larger detuning, in any polarizations, blue shifts are much smaller than predicted by the dressed exciton model. We present a tentative model, which is an extended dressed exciton model including the excitonic molecule and unbound biexcitons phenomenologically. The model reproduces the experimental result quite well.

DOI: 10.1103/PhysRevB.69.155201 PACS number(s): 71.35.−y, 42.50.−p, 42.65.−k, 78.47.+p

I. INTRODUCTION

Since early reports,[1–3] the optical Stark effect of excitons has been understood phenomenologically by the *dressed exciton picture*, which is the analog of the dressed atom.[4] The exciton is assumed to be a two-level system, and the shift is described as, for weak light,

$$\delta\omega_X = -|\mu_X E|^2/(\omega_p - \omega_X), \tag{1}$$

where ω_X, $\delta\omega_X$, and μ_X represents, the energy, the energy shift, and the transition dipole moment of the exciton, respectively. The frequency and the electric field of light are expressed by ω_p and E, respectively. The numerator represents the light-exciton coupling, and the denominator represents the detuning. The picture is strongly supported by clear observations in the GaAs quantum well that the shift was proportional to the light intensity[3] and to the inverse of the detuning[3] and that the effect was independent of light polarizations.[2]

On the other hand, these observations stimulated a number of theoretical works, which focus on many-body aspects of the excitonic optical Stark effect. First-principle theories, which are derived from elementary interactions between conduction-band electrons and valence-band holes, based on the Hartree-Fock approximation,[5,6] the Green's function technique,[7] and the perturbation[8,9] have been presented. Distinctive features in the excitonic system, all of which originate from the Coulomb interaction, have been pointed out, for instance, the excitonic enhancement,[5] the stronger detuning dependence,[8,10,11] and the oscillator strength change.[5,12,13]

Predicted peculiarities, however, have been detected in only a few experiments.[11,14–16] The excitonic redshift was observed, by the pump-probe spectroscopy, in the CuCl crystal[14] in the colinear configuration and in the InGaAs quantum well in the opposite-circular configuration.[15] The former was explained to be due to the coupling between the exciton and the excitonic molecule, as the pump energy was just below its resonance.[8,9,17] The latter was explained to be due to the four-particle (two electrons and two holes) correlation and the Coulomb memory effect, in comparison with the one-dimensional full $\chi^{(3)}$ calculation.[18] As for the detuning dependence, in order to investigate contributions of the heavy hole and the light hole in the InGaAs quantum well, Brick *et al.*,[11] compared differential spectra with three different detunings, and examined the detuning dependence with the theoretical support. However, these observations are not enough to understand the whole picture of the excitonic optical Stark effect, especially in the system with strong Coulomb interaction, where the exciton-exciton interaction may play dominant roles.[19] Systematic measurement of the shift with continuously changing detuning in the scale of the exciton Rydberg is desirable.

In this paper, we report the systematic measurement of the detuning dependence of the excitonic optical Stark shift in an inorganic-organic layered semiconductor, $(C_6H_5C_2H_4NH_3)_2PbI_4$.[20,21] In this semiconductor, the exciton is confined in inorganic layers; organic layers work as barriers for the confinement. This substance is suitable for studying many-body aspects of the excitonic optical Stark effect for two reasons. One is the strong effective Coulomb interaction between photogenerated carriers, which enhances the correlation between excited quasiparticles, such as the exciton. The exciton Rydberg is 220 meV. The binding energy of the excitonic molecule is 40–45 meV.[22–24] The other is the strong exciton-light coupling, which is the source of the prominent Stark shift. We show the polarization dependence and the detuning dependence of the excitonic Stark shift with the continuously changing detuning in a range of 350 meV below the exciton resonance. Strong biexcitonic contributions are clarified. These results are compared with the extended dressed exciton model which includes biexcitonic levels. Although the model is phenomenological and a more careful treatment might be necessary for precisely examining physical sources of the Stark effect, the model reproduces the experimental data surprisingly well.

II. EXPERIMENT

$(C_6H_5C_2H_4NH_3)_2PbI_4$ was prepared by the synthetic method.[20] The sample for the measurement was a polycrystalline film prepared by spin coating from the solution. The sample was placed on the cold finger of a cryostat and was cooled to about 10 K.

The Stark shift of the exciton was measured with the subpicosecond pump-probe technique in the transmission geometry.[22] Two split-off beams of the output from a 1 kHz Ti-sapphire regenerative amplifier were used for generating the pump and probe lights. The pump light was generated with an optical parametric amplifier, which covers all the wavelength range in the experiment. As the probe light, the white light continuum was generated by the self-phase-modulation in a water cell. Polarizations of both pump and probe lights were operated with achromatic wavelength plates in four configurations: cocircular $(\sigma^+\sigma^+)$, opposite circular $(\sigma^+\sigma^-)$, colinear (xx), and cross-linear (xy). The overlap of pump and probe spots on the sample was checked with a video telescope. Spot sizes were measured with a pinhole fixed on a micrometer stage. The cross correlation and the zero delay time $(\tau_d=0)$ between pump and probe pulses were measured by the sum-frequency generation with a β-BaB_2O_4 crystal, folding the beam paths in front of the cryostat by a mirror. The cross correlation was typically 230 fs in the full width at half maximum. The intensity and the pulse width of the pump light were measured with a photodiode powermeter and an autocorrelator, respectively. The angle between pump and probe beams was 11°. Since the delay time is a function of the probe wavelength, retrieved spectra were numerically corrected after measurements. In this paper, we report the data at $\tau_d=0$, unless otherwise noted.

III. RESULT AND DISCUSSION

The solid line in Fig. 1 shows the linear absorption spectrum. Two absorption bands at 2.350 and 2.395 eV are assigned to $1s$ excitons which are split because the unit cell consists of two formula units.[23] The intensity ratio of higher to lower bands is independent of polarizations and is estimated to be about 0.35 by spectral deconvolution. In this paper, we focus on the exciton at 2.350 eV. We consider that the exciton at 2.395 eV is not considerably influencing to the excitonic shift of our interest, due to the small oscillator strength and the energy separation.[25]

Photoinduced spectrum changes, as examples, are drawn by dashed and dash-dotted lines for $\sigma^+\sigma^+$ and $\sigma^+\sigma^-$, respectively. The pump photon energy is 2.298 eV. It is observed that the exciton peak prominently blue shifts in $\sigma^+\sigma^+$, whereas it red shifts in $\sigma^+\sigma^-$. Although not shown in the figure, we have confirmed that the red shift is also observed in xx, while not in xy.[14] Therefore, it is now found that the polarization selection rule for this red shift is the same as that for the two-photon generation of the excitonic molecule.[17] Moreover, since the pump detuning is only slightly more than the binding energy of the excitonic molecule by 7 meV, the red shift is qualitatively explained to be due to the coupling between the exciton and the excitonic molecule.

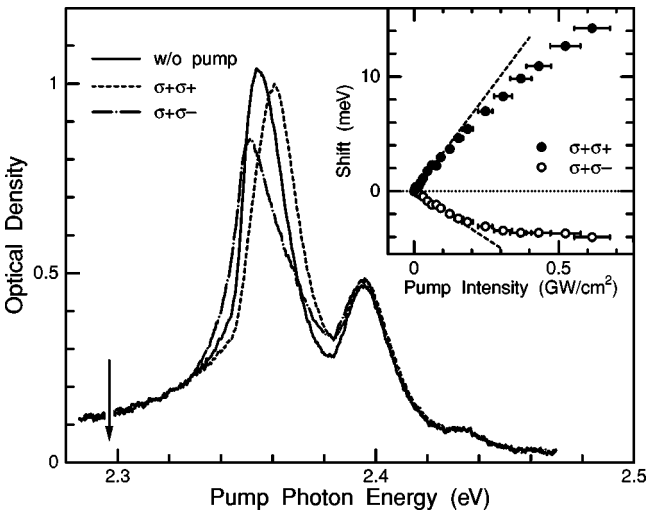

FIG. 1. Absorption spectra with and without the pump. The pump photon energy is 2.298 eV, as shown by the arrow. The inset shows the pump intensity dependence of the shifts under the same conditions.

We note, in the case that the pump light and the exciton absorption band have spectral overlap to generate real excitons, not only the prominent shift at $\tau_d\approx 0$ but also a weak long lived ($\approx$10 ps) blue shift are observed. The long lived blue shift is ascribed to the interaction between decohered excitons.[26] While, we ascribe the shift at $\tau_d=0$ fully to the Stark shift, assuming that the excitons at $\tau_d\approx 0$ are conserving the initial coherence. The cross correlation is much less than the dephasing time of 500 fs.[27]

The inset of Fig. 1 shows the intensity dependence of the shift.[28] In both $\sigma^+\sigma^+$ and $\sigma^+\sigma^-$, the shifts are sublinear to the intensity. The sublinear dependences are essential for the dressed exciton model, $\delta\omega_X=2\sqrt{|\mu_X E|^2+(\omega_p-\omega_X)^2}$, which gives the linear dependence in the low intensity limit, i.e., $|\mu_X E|/|\omega_p-\omega_X|\ll 1$, as is given by Eq. (1). The obtained result shows the linear dependence up to 1×10^8 W/cm^2. We confine ourselves into this linear regime, in the following.

Figure 2 shows the detuning dependence of the shift in two overlapping ranges. The ordinate represents the shift normalized by the pump intensity, assuming the linear regime. Curves are calculated with the model explained below. Note that scales of both horizontal and vertical axes are different in between (a) and (b).

It is clearly found that, in $\sigma^+\sigma^-$ and xx, the shift changes its sign at 2.300 ± 0.002 eV and is enhanced at both the higher and lower sides.[29] This feature is the obvious evidence of the resonance of the pump light to the transition between the exciton and the excitonic molecule.[14,8] As is slightly different from the prediction,[9] we find that the detuning energies (50 meV) for these crossovers are larger than the exact value of the binding energy (40–45 meV) of the excitonic molecule. This difference is explained to be due to small competing contributions for the blue shift discussed later. Whereas, in $\sigma^+\sigma^+$ and xy, the blue shift monoto-

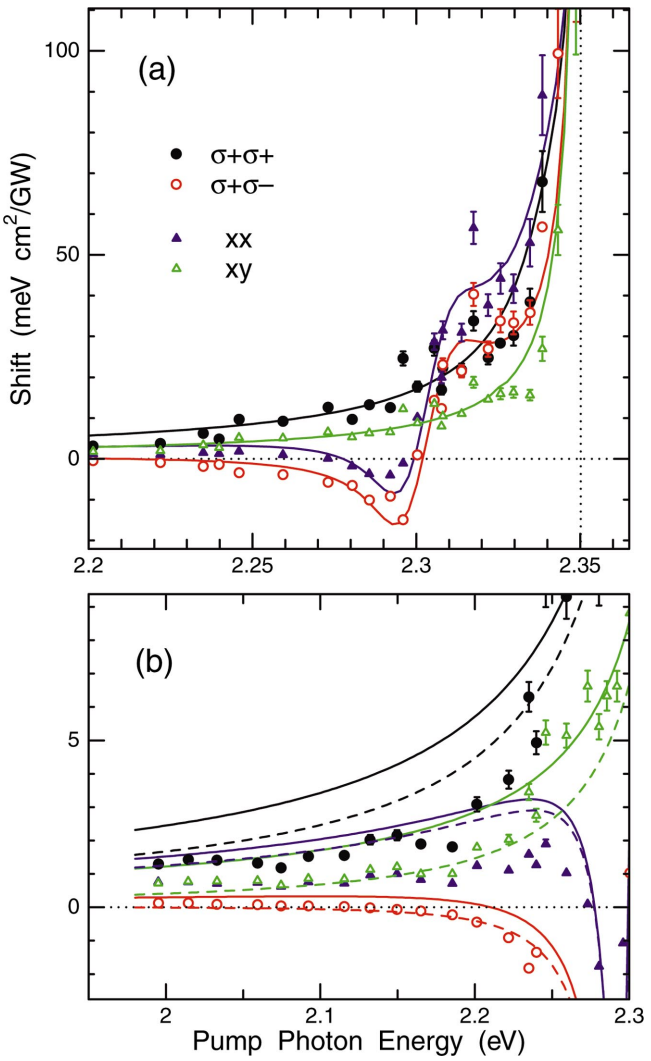

FIG. 2. (Color) Pump photon energy dependence of the shift in $\sigma^+\sigma^+$ (black), $\sigma^+\sigma^-$ (red), xx (blue), and xy (green) for two overlapping ranges. Solid and dashed curves are drawn according to the model without and with unbound biexcitons, respectively. The vertical dotted line shows the exciton resonance. In (b), data are plotted in the magnified vertical scale in the range less than 2.3 eV. The horizontal scale is reduced.

nously decreases for larger detuning.

Moreover, quantitative relations are pointed out. The blue shift in $\sigma^+\sigma^+$ is about twice of that in xy. The shift in xx is larger than that in $\sigma^+\sigma^-$ to the positive direction. The range for the red shift in $\sigma^+\sigma^-$ is rather large compared with that in xx: from 2.13 to 2.30 eV in $\sigma^+\sigma^-$ and from 2.28 to 2.30 eV in xx. These polarization dependences in detail are clearly understood as a result of competing factors, which are described by the model discussed below.

Although the detuning dependence has been derived analytically by Combescot,[8] it is not possible to concretely calculate it without knowing the biexcitonic wave functions. Another way is the numerical calculation based on the first-principle kinetic equations.[30] But, we believe that it is not straightforward to apply these theories to our system, because the effective Coulomb potential is strong and not conventional in this substance due to the dielectric confinement effect.[21,26] Thus, in this paper, we try to understand the experimental result with a tentative model, where the biexcitonic states are phenomenologically introduced to the simple dressed exciton model.

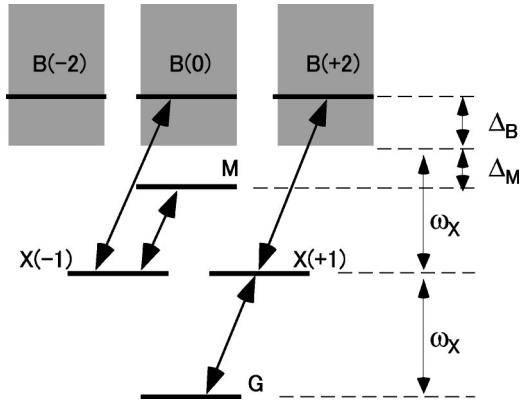

FIG. 3. Levels for the extended dressed exciton model. G, $X(\pm 1)$, M, and $B(0,\pm 2)$ represent the ground state, the exciton, the excitonic molecule, and unbound biexcitonic states, respectively. Gray zones show distributions of unbound biexcitonic states. Arrows between levels show nonzero coupling by the σ^+-polarized light.

We shall postulate several levels for the exciton system, as is schematically shown in Fig. 3. The ground state, the excitons, the excitonic molecules, and unbound biexcitons are represented as G, $X(\pm 1)$, M, and $B(0,\pm 2)$, where the numbers in parentheses represent the component of the total angular momenta (J_z) along the light axis.

For modeling the system, we consider only the following states on the assumption of low light intensity. Because of the momentum conservation rule, neglecting light momenta, only excitons and biexcitons with zero translational momentum are allowed for optical coupling. Additionally for unbound biexcitons, the relative momentum between two constituent excitons must be zero. But, relative momenta are not quantum numbers for the eigenstates; the oscillator strength from the exciton is shared by continuously distributed states of unbound biexcitons. To simplify this complexity, we postulate that this group of states can be replaced by a representative level B.

Among levels mentioned above, G and $X(+1)$, $X(-1)$ and M, $X(-1)$ and $B(0)$ and $X(+1)$, and $B(+2)$ couple by the σ^+-polarized light according to the conservation rule of angular momenta. We express transition moments for these transitions by $\mu_{G,X}$, $\mu_{X,M}$, $\mu_{X,B(0)}$, and $\mu_{X,B(2)}$, respectively. Because a biexciton transition is a process of adding one more exciton to the system, we assume $\mu^2_{X,B(\pm 2)} = \mu^2_{X,M} + \mu^2_{X,B(0)} = \mu^2_{G,X}$.[31] Then, depending on the polarizations, the change of the G-X transition energy is expressed as

$$\delta\omega_{++} = 2\frac{|\mu_{G,X}E|^2}{\omega_X - \omega_p} - \frac{|\mu_{X,B(2)}E|^2}{\omega_X + \Delta_{B(2)} - \omega_p}, \qquad (2)$$

$$\delta\omega_{+-}=\frac{|\mu_{G,X}E|^2}{\omega_X-\omega_p}-\frac{|\mu_{X,M}E|^2}{\omega_X-\Delta_M-\omega_p}-\frac{|\mu_{X,B(0)}E|^2}{\omega_X+\Delta_{B(0)}-\omega_p}, \tag{3}$$

$$\delta\omega_{xx}=2\frac{|\mu_{G,X}E|^2}{\omega_X-\omega_p}-\frac{|\mu_{X,M}E|^2}{\omega_X-\Delta_M-\omega_p}-\frac{|\mu_{X,B(0)}E|^2}{\omega_X+\Delta_{B(0)}-\omega_p}-\frac{1}{2}\frac{|\mu_{X,B(2)}E|^2}{\omega_X+\Delta_{B(2)}-\omega_p}, \tag{4}$$

$$\delta\omega_{xy}=\frac{|\mu_{G,X}E|^2}{\omega_X-\omega_p}-\frac{1}{2}\frac{|\mu_{X,B(2)}E|^2}{\omega_X+\Delta_{B(2)}-\omega_p}, \tag{5}$$

where $-\Delta_M$ (<0) and $\Delta_{B(0,2)}$ (>0) represent the correlation energy for *M* and *B*, respectively. Expressions for *xx* and *xy* are obtained via decomposing light and exciton polarizations in the circular basis of polarizations.

Solid and dashed curves in Figs. 2(a) and 2(b) show best fits in the range of 2.3 ± 0.05 eV, without and with *B*, respectively. We note that the calculation was actually performed by numerical diagonalization of the model Hamiltonian,[32] which is equivalent to Eqs. (2)–(5) in the low intensity limit. Linewidths of transitions and the pump light are also introduced phenomenologically.[33] Free adjustable parameters are $\mu_{G,X}/\mu_{X,M}$ and $\Delta_{B(0,\pm2)}$ only; other quantities are restricted by values obtained experimentally.[34] Dashed curves are not drawn in (a), because they almost overlap with the corresponding solid curves. We shall first discuss the fit without *B*. The fit with *B* is discussed later for explaining the quantitative deviation of the fit without it.

The numerical result reproduces the experimental result quite well, especially in the range larger than 2.2 eV, as is seen in Fig. 2(a). Data in four polarizations are simultaneously fit. In $\sigma^+\sigma^-$ and *xx*, the resonance enhancement and the red shift–blue shift crossover are reproduced well. These are mainly caused by the *X*-*M* coupling given by the second terms in Eqs. (3) and (4), the denominators of which give the resonance and change their signs at $\omega_X-\Delta_M$. The crossover energies are slightly lower than the exact *X*-*M* resonance by ca. 5 meV, because the *G*-*X* coupling given by the first term in the Eqs. (3) and (4) gives small positive contributions.

The blue shift–red shift crossover is also seen at larger detuning in $\sigma^+\sigma^-$ and *xx*: at 2.13 eV and 2.27 eV in experiment, while at 2.22 eV and 2.28 eV by the model, respectively. According to the model, these crossover energies are also determined by the balance between *G*-*X*, *X*-*M*, and *X*-*B* couplings. Although the crossover energy by the model without *B* does not fit well in $\sigma^+\sigma^-$, the fit is much improved by including the *X*-*B* coupling, as is seen below.

Systematic relations between different polarizations found in the experimental result are also reproduced. The blue shift in $\sigma^+\sigma^+$ is the twice of that in *xy*, i.e., $\delta\omega_{++}=2\,\delta\omega_{xy}$. The shift in *xx* is always larger than that in $\sigma^+\sigma^-$, of which difference is given as $\delta\omega_{xx}-\delta\omega_{+-}=\delta\omega_{xy}$. These relations originate from the polarization selection rule for biexcitons and stand regardless of the parameter values.

For the range far from the *X*-*M* resonance, the fit of the numerical result without *B* is quantitatively not good, as shown by solid curves in Fig. 2(b). The calculated shifts in all polarizations are roughly twice those of the experimental data in the range less than 2.2 eV. This discrepancy is considerably large, considering the experimental resolution. Because shifts should be proportional to the inverse of the detuning for $\sigma^+\sigma^+$ and *xy* in the model without *B*, the discrepancy represents the stronger dependence, i.e., $\delta\omega_{++,xy}\propto|\omega_p-\omega_X|^{-\alpha}$ with $\alpha>1$, although the value of α cannot be determined accurately with our experimental resolution. Such strong dependence was theoretically derived as a result of the Coulomb interaction[8,12] or more concretely the exciton-exciton interaction,[10] but this has been suggested in only a few experiments.[11,16]

Including *B* to the model, the numerical result gives a rather improved fit, as shown by the dashed curves in Fig. 2(b). Blue shifts in all configurations become significantly suppressed by the *X*-*B* coupling which lowers the energy of *X*. Based on this improved fit, we conclude the unbound biexcitonic states as the main source of the suppressed blue shift in the range less than 2.2 eV.[25] We note that the *X*-*B* coupling is less important for smaller detuning; the fits with and without *B* are really identical, because the *G*-*X* coupling and the *X*-*M* coupling are resonantly enhanced and are predominant.

Our result, which strongly implies the contribution of the unbound biexcitons, is in contrast to the numerical result reported by Brick *et al.*[11] Based on the semiconductor Bloch equation approach, they show, the shift in the InGaAs quantum well is dominated by the Pauli exclusion principle and is scarcely contributed by the Coulomb interaction in $\sigma^+\sigma^+$ where no stable excitonic molecule exists. This difference is most likely explained to be due to the difference in the strength of the effective Coulomb interaction. The exciton Rydberg in the inorganic-organic layered semiconductor is about 20 times as large as that in the InGaAs quantum well. Thus, the Coulomb contribution seems to be exaggerated in our substance.

IV. CONCLUSION

We have clarified the detuning dependence of the excitonic optical Stark shift in the semiconductor where the effective Coulomb interaction is strong. The enhancement and the red shift–blue shift crossover are observed at a detuning of 50 meV in $\sigma^+\sigma^-$ and *xx*. These are ascribed to the resonant contribution of the excitonic molecule. Blue shifts in any polarization in the range less than 2.2 eV is found to be much smaller than predicted by the simple dressed exciton model. This suppression of blue shifts are ascribed to the red shift contribution by the unbound biexcitons. An extended dressed exciton model which includes the excitonic molecule and the unbound biexcitons is presented to show the close reproduction of the experimental result.

ACKNOWLEDGMENT

The work was partly supported by the Grant-in-Aid for Young Scientists (B) (Grant No. 15740189) of MEXT, Japan.

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[33] We diagonalized the model Hamiltonian to determine the energy and intensity of transitions. Then, a spectrum was calculated on the assumption that all transitions have Gaussian linewidths of $\sigma = 7$ meV. The pump light was assumed to be monochromatic. Obtained spectra were used for determining the peak energies, in the same way as experimentally observed spectra.

[34] Best fits without and with B are demonstrated by $\mu_{X,M}^2/\mu_{G,X}^2 = 0.65$ and 0.6, respectively. Regardless of B, $\omega_X = 2.350$ eV and $\Delta_M = 49$ meV are used. With B, $\Delta_{B(0,\pm 2)} = 100$ meV is used, but the result is not sensitive for more than 50 meV.

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