Observation of Autler-Townes Splitting of Biexcitons in CuCl

Ryo Shimano and Makoto Kuwata-Gonokami*
Department of Applied Physics, Faculty of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan
(Received 18 October 1993)

Wave-vector–selective ac Stark effect of the Γ_1-biexciton state is clearly observed in the steady state regime. The biexciton line splits into two lines with resonant pumping between Z_3 exciton polariton and the biexciton state. From the splitting energy, we have obtained the relevant dipole moment of 4.2e Å. Observed energy of the dressed state coincides with the value derived from the model with strict energy and quasimomentum selection rule. This implies that the dressed state is established with the coherent mixing of delocalized Bloch states.

PACS numbers: 71.35.+z, 42.50.Hz, 71.36.+c

In atomic systems, the concept of the dressed atom has been established both theoretically and experimentally [1]. The dressed atom state has received a great deal of attention in several applications such as manipulation of kinetic motion of atoms [2,3] and lossless optical conversion and lasers without inversion [4,5]. On the other hand, the development of tunable short pulsed lasers enables us to study the dressed states in semiconductors. In particular, the ac Stark effects of excitons have been studied extensively after the demonstration of a very quick blueshift of the exciton state in the GaAs quantum well (QW) under intense excitation below the exciton resonance [6]. To examine the similarities and dissimilarities between exciton and atomic systems, we have to take the following two features of excitons. One is the many-body effect caused by the Coulomb and exchange interaction between electrons and holes. The second is the effect of the delocalized nature of the exciton state which is in strong contrast with the atomic system where the electron wave function is very small and localized. In crystals, every electronic state is the eigenstate of the displacement operator because of the translational symmetry and its wave function is delocalized. Such a state is called a Bloch state. It is important to know how the dressed states are established in the case of Bloch states, where every optical transition obeys the energy and quasimomentum conservation rules. To examine this effect, we need to measure the wave-vector dependent change of the Bloch states under resonant optical excitation. In semiconductors, however, resonant optical pumping causes strong incoherent processes because of the scattering between excitons and photoexcited carriers. Therefore, most of the previous experiments have been performed in the ultrafast time domain [7–9], sacrificing spectral resolution. Only a few experiments in the steady state are reported on the exciton ac Stark effects [10,11] and no systematic study on the quasimomentum space has been performed. In this Letter, we report a clear demonstration of the semiconductor dressed states where the wave-vector–dependent coupling is essential. We present the experimental evidences of the Rabi splitting of the biexciton line caused by the coherent optical mixing with the exciton polariton state.

The biexciton state, where two Wannier excitons are bound by the exchange interaction between electrons, has been observed in various semiconductors. Nonlinear optical processes associated with the formation of biexcitons can be described by the three level diagram as shown in Fig. 1(a). If the pump beam frequency is tuned to the transition between exciton state ($|ex\rangle$) and biexciton state ($|bi\rangle$), the two excited states are mixed to create dressed states as shown in Fig. 1(a). This corresponds to the Autler-Townes (AT) effects [12] in atomic systems. In this scheme, the pump beam can be set below the exciton absorption band; thus we can observe the coherent effect in the steady state regime. Three level ac Stark

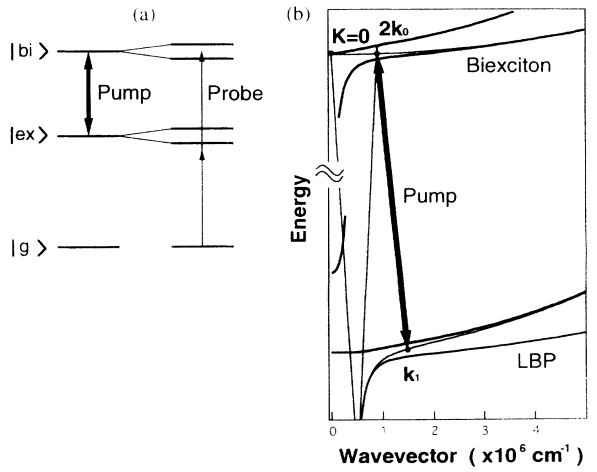

FIG. 1. (a) Schematic level diagram of the exciton and biexciton system. The exciton and biexciton states are coherently mixed by the resonant pump field. The resulting dressed states are probed by two-photon polarization (TPP) spectroscopy. (b) Calculated Autler-Townes splitting taking into account the exciton-polariton effect with spatial dispersion. The pump field mixes the lower branch polariton (LBP) with the biexciton branch. TPP spectroscopy probes the $\mathbf{K} = 2\mathbf{k}_0$ biexciton state.

0031-9007/94/72(4)/530(4)$06.00

effects have been reported by Fröhlich *et al.* in their pioneering work on Cu_2O [13] and later also on the GaAs QW [14], where the levels are single exciton states. In the exciton-biexciton three level system, the splitting is proportional to the Rabi frequency, $\Omega_R = \mu_{bx} E_{pump}$, where μ_{bx} is the dipole transition matrix element between exciton and biexciton states and E_{pump} is the internal electric field vector of the pump beam. Because of the large Bohr radius of biexcitons, the dipole moment μ_{bx} is large; therefore the splitting becomes very large.

In the actual optically allowed exciton transition, we have to take into account the polariton effect. The exciton polariton is the eigenmode of the total system of photons and excitons with strong linear coupling between them. Figure 1(b) represents the AT effect on the base of the polariton picture including spatial dispersion effects of the exciton polariton and the biexciton states. In this scheme, optical transitions occur keeping the energy and quasimomentum conservation rule. The pump beam is incident on the rear surface of the sample. We probe the biexciton state at $\mathbf{K}=2\mathbf{k}_0$ with a two-photon transition, where $\mathbf{k}_0$ is the wave vector of the probe beam. As long as the pump field is uniform, coherent mixing occurs between two states with the same energy and the quasimomentum; one is the biexciton state at $\mathbf{K}=2\mathbf{k}_0$ and the other is the combined state of a pumping photon (a polariton) with wave vector $-\mathbf{k}_p$ and exciton polariton with wave vector $\mathbf{k}_L = 2\mathbf{k}_0 + \mathbf{k}_p$. The eigenenergy of dressed biexciton states is expressed as follows [15,16]:

$$\Lambda_{\pm} = \frac{\Omega_1(\mathbf{K}) + \Omega_2(\mathbf{K}) \pm \sqrt{[\Omega_1(\mathbf{K}) - \Omega_2(\mathbf{K})]^2 + \Omega_R^2}}{2}, \tag{1}$$

with

$$\Omega_1(\mathbf{K}) = \Omega_B(0) + \frac{\hbar^2 K^2}{2M}, \tag{2}$$

$$\Omega_2(\mathbf{K}) = \Omega_{EP}(\mathbf{K} + \mathbf{k}_p) + \Omega_{EP}(\mathbf{k}_p), \tag{3}$$

$$\Omega_R = \mu_{bx} E_{pump}. \tag{4}$$

Here, $\Omega_B(\mathbf{K})$ and $\Omega_{EP}(\mathbf{k})$ are the energies of the biexciton and exciton polariton, M is the effective mass of a biexciton, μ_{bx} is the dipole moment between a biexciton state and an exciton state, and E_{pump} is the internal electric field amplitude of the pump beam. The formation of dressed states can be proved as the splitting of the biexciton resonances in the two-photon absorption (TPA) spectrum. The coherent coupling also causes the modification of eigenenergies of exciton polariton states as shown in Fig. 1(b). The latter phenomenon is nothing but the induced change of the polariton dispersion which is known as the "polariton renormalization effect" [17,18]. Several experimental evidences of this effect have been reported, although indirectly, such as a photoinduced birefringence [19], a nonlinear shift of hyper-Raman scattering [20], and, more recently, a redshift of the exciton band in subpicosecond pump and probe experiments [21].

The Γ_1-biexciton state in CuCl, the lowest bound state of two Z_3 excitons, has been studied extensively by two-photon absorption [22–24] and hyper-Raman scattering [20,25,26]. The Γ_1-biexciton state is a suitable system for the demonstration of the AT effect because of the following features. First, it has a large binding energy, 32 meV; we can set the pump beam frequency far below the exciton absorption band and thus the coherent effects occur predominantly even in steady state excitation. The second feature is the very narrow linewidth of the Γ_1-biexciton state. At liquid helium temperature, the linewidth of the biexciton state is less than 20 μeV because of very small acoustic phonon coupling [27]. Such narrow linewidth is appropriate for a precise measurement of the energy shift.

High purity platelet-shaped CuCl single crystals of thickness 2 μm to 15 μm are cooled in a helium gas flow-type cryostat. To monitor the biexciton resonance energy, we use two-photon polarization (TPP) spectroscopy whose principle was described in Ref. [28]. Two dye lasers, with pulse durations of 5 nsec and linewidths of 20 μeV (FWHM), are excited by one excimer laser (Lambda Physik LPX120i). One dye laser (ω_1) is split into two beams; one is linearly polarized (beam 1 with 1 kW/cm^2) and the other is circularly polarized (beam 2 with intensity 10 kW/cm^2). At the two-photon resonance of the biexciton, $\omega_1 = \Omega_B/2$, the circularly polarized pump beam causes a nonlinear optical activity and the probe polarization changes. Thus we can probe the biexciton two-photon resonance sensitively as the positive signal with crossed analyzer. An etalon plate with 50 GHz free spectral range is used to monitor the probe frequency. A second dye laser is used as a pump beam (beam 3) to couple the exciton polariton state and the biexciton state. We change the pump beam intensity from 10 kW/cm^2 to 1 MW/cm^2. Beams 1 and 2 impinge on the sample from the same side and probe the biexciton state around $\mathbf{K}=2\mathbf{k}_0$, where $\mathbf{k}_0$ is the polariton wave vector at the two-photon resonance of the biexciton.

First, we set the pump beam (beam 3) energy at 3.17009 eV, the exact resonance between exciton polariton ($\mathbf{k}_L = 2\mathbf{k}_0 + \mathbf{k}_p$) and biexciton ($\mathbf{K}=2\mathbf{k}_0$). Figure 2 shows the two-photon polarization spectra around the biexciton line, 3.1860 eV, for various pump powers. We clearly observed the splitting of the biexciton line whose splitting energy becomes large as the pump beam power increases. These two peaks correspond to the dressed states. From the splitting energy, we can evaluate the value of μ_{bx}. We fit the data to the theoretical calculation of Eq. (1) and obtain good agreement between the experiment and the theory with the parameter $\mu_{bx} = 4.2e$ Å. The corresponding value of the oscillator strength of the exciton-biexciton transition, f_m, is 15 which is much larger than that of the Z_3 1s exciton, $f_{ex}=5.9 \times 10^{-3}$. The large radius of the biexciton wave function causes

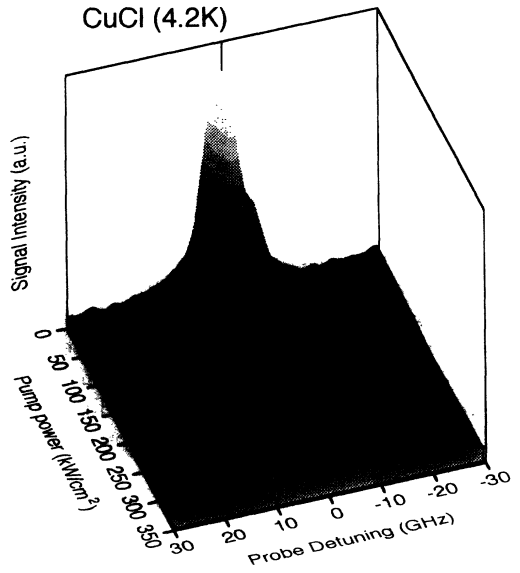

FIG. 2. TPP spectra around the biexciton line, 3.186 eV, for various pump powers. The pump detuning is almost zero.

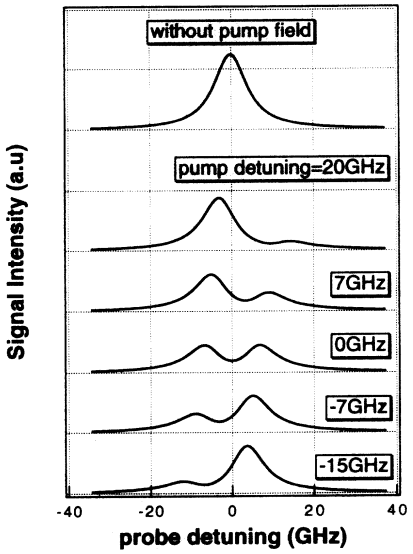

FIG. 3. Calculated TPP spectra around the biexciton line for various pump laser detunings. The biexciton dephasing rate Γ_{mg} is set to $\Omega_R/5$.

this enhancement. The enhancement factor can be expressed as [29,30]

$$\frac{f_m}{f_{ex}} = \frac{2}{v}\left[\int g(R)\, dR\right]^2 , \tag{5}$$

where $g(R)$ is the wave function of the relative motion of the two excitons in a biexciton and $v = (0.54\ \text{nm})^3/4$ is the volume of the primitive unit cell. Our result gives the value of $f_m/f_{ex} = 2.5 \times 10^3$. With the weak two-photon excitation condition, Akiyama *et al.* measured the radiative decay rate of the biexciton and reported a value of 50 ps [31] which leads to $f_m/f_{ex} = 3.3\times10^3$. This short radiative lifetime is also supported by the pump and probe experiments [32] and the emission decay measurement under incoherent excitation of biexcitons [33]. This value is consistent with our estimated value of f_m/f_{ex} from the ac Stark splitting. This agreement assures that a rapid spontaneous radiative decay process is the main decay channel of the biexcitons in CuCl.

To examine the pump photon energy dependence, we study the two-photon polarization spectra by changing the pump photon energy around the exciton-biexciton transition. Figure 3 shows the calculated spectra for various pumping energies. The crossing point of the two modes, i.e., the solution of energy and momentum conservation equations at zero pump field limit, varies with the pump energy. For negative detuning ($\hbar\omega_{\text{pump}} < \Omega_P^0$), the crossing point exists at the wave-vector region larger than the probing region ($\sim 2\mathbf{k}_0$). In this case the upper state $|\Lambda_+\rangle$ is the biexcitonlike state and has a stronger two-photon absorption amplitude than the lower branch $|\Lambda_-\rangle$. For positive detuning ($\hbar\omega_{\text{pump}} > \Omega_P^0$), the situation becomes contrary. Figure 4 shows the experimental results. The basic features, the relative intensities of the upper and lower branches and the energy positions, fit well with the calculation. In the experimental data, we observe an additional line at the low energy side. The spectral position of this line is independent of the pump field energy and power. From the precise analysis, the line is assigned to the biexciton state at $\mathbf{K}$=0. A part of beam 2 is reflected at the rear surface and induces the transition into the $\mathbf{K}$=0 state; thus we observe this line. Because of the energy and momentum conservation rule, the $\mathbf{K}$=0 biexciton state cannot couple with the pumping

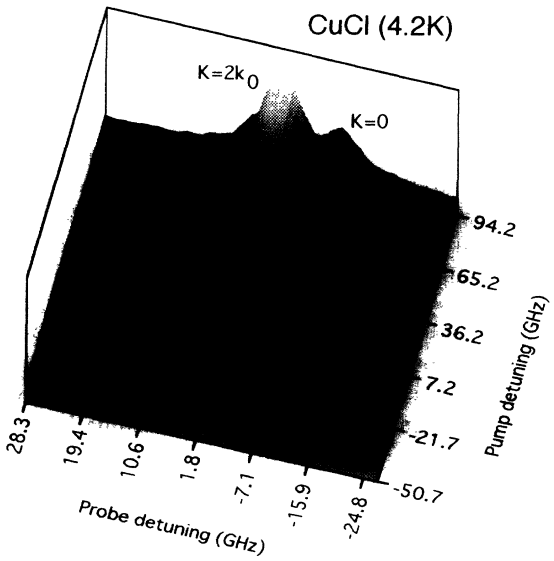

FIG. 4. Observed TPP spectra around the biexciton line for various pump laser detunings.

field of beam 3. This is the reason why the $\mathbf{K}=0$ state does not change in Fig. 4.

We obtained a clear experimental evidence of the Autler-Townes splitting of biexcitons in CuCl. We have proved that coherent mixing of exciton polaritons and biexcitons occurs under strict energy and quasimomentum selection rules. This is the first observation of dressed delocalized Bloch states. We compare the experiment with the calculation and obtained the value of the giant oscillator strength of the biexciton, $f_m/f_{ex} = 2.5 \times 10^3$, which is consistent with the reported radiative decay time. The observed phenomena, namely, a modification of the eigenenergy of the biexciton state with the radiation field, provides us with a new method to control the kinetic motion of biexcitons by using the spatially inhomogeneous radiation field. This will provide us with a key technique to confine or manipulate biexcitons into the quantum degenerate regime.

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

* To whom correspondence should be adressed.

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1992).
[2] G. Grynberg, B. Lounis, P. Verkek, J.-Y. Courtois, and C. Salomon, Phys. Rev. Lett. **70**, 2249 (1993).
[3] A. Hemmerich, C. Zimmermann, and T. W. Hänsch, Europhys. Lett. **22**, 89 (1993).
[4] S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989).
[5] Y. Zhu, Q. Wu, S. Morin, and T. W. Mossberg, Phys. Rev. Lett. **65**, 1200 (1990).
[6] A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoç, Phys. Rev. Lett. **56**, 2748 (1986).
[7] N. Peyghambarian, S. W. Koch, M. Lindberg, B. Fluegel, and M. Joffre, Phys. Rev. Lett. **62**, 1185 (1989).
[8] B. Fluegel, N. Peyghambarian, G. Olbright, M. Lindberg, S. W. Koch, M. Joffre, D. Hulin, A. Migus, and A. Antonetti, Phys. Rev. Lett. **59**, 2588 (1987).
[9] W. H. Knox, D. S. Chemla, D. A. B. Miller, J. B. Stark, and S. Schmitt-Rink, Phys. Rev. Lett. **62**, 1189 (1989).
[10] C. Klingshirn, Semicond. Sci. Technol. **5**, 457 (1990); **5**, 1006 (1990).
[11] I. Akai, T. Karasawa, T. Komatsu, and Y. Kaifu, Phys. Status Solidi (b) **150**, 635 (1988).
[12] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).
[13] D. Fröhlich, A. Nöthe, and K. Reimann, Phys. Rev. Lett. **55**, 1335 (1985).
[14] D. Fröhlich, K. Reimann, and R. Wille, J. Lumin. **38**, 235 (1987).
[15] M. Kuwata-Gonokami and R. Shimano, in *Proceedings of the Tenth International Conference on Laser Spectroscopy, France, 1991*, edited by M. Ducloy, E. Giacobino, and G. Camy (World Scientific, Singapore, 1992), p. 156.
[16] For theoretical references on the biexciton ac Stark effect see, e.g., E. Hanamura, Phys. Rev. B **44**, 8514 (1991); A. L. Ivanov and H. Haug, Phys. Rev. B **48**, 1490 (1993); M. Combescot and R. Combescot, Phys. Rev. B **40**, 3788 (1989).
[17] V. May, K. Henneberger, and F. Henneberger, Phys. Status Solidi (b) **94**, 611 (1979).
[18] R. März, S. Schmitt-Rink, and H. Haug, Z. Phys. B **40**, 9 (1980).
[19] M. Kuwata, T. Mita, and N. Nagasawa, Solid State Commun. **40**, 911 (1981); M. Kuwata and N. Nagasawa, J. Lumin. **31**, 867 (1984).
[20] T. Itoh and T. Suzuki, J. Phys. Soc. Jpn. **45**, 1939 (1978).
[21] D. Hulin and M. Joffre, Phys. Rev. Lett. **65**, 3425 (1990).
[22] G. M. Gale and A. Mysyrowicz, Phys. Rev. Lett. **32**, 727 (1974).
[23] N. Nagasawa, N. Nakata, Y. Doi, and M. Ueta, J. Phys. Soc. Jpn. **39**, 987 (1975).
[24] Vu Duy Phach, A. Bivas, B. Hönerlage, and J. B. Grun, Phys. Status Solidi (b) **84**, 731 (1977); R. Levy, B. Hönerlage, and J. B. Grun, Phys. Rev. B **19**, 2326 (1979).
[25] B. Hönerlage, A. Bivas, and Vu Duy Phach, Phys. Rev. Lett. **41**, 49 (1978).
[26] T. Mita, K. Sotome, and M. Ueta, Solid State Commun. **33**, 1135 (1980).
[27] M. Kuwata-Gonokami, R. Shimano, J. Iwamatsu, H. Akiyama, T. Kuga, and M. Matsuoka, Phys. Status Solidi (b) **159**, 347 (1990).
[28] M. Kuwata, J. Phys. Soc. Jpn. **53**, 4456 (1984).
[29] A. A. Golouin and E. I. Rashba, Pis'ma Zh. Eksp. Teor. Fiz. **17**, 690 (1973) [JETP Lett. **17**, 478 (1973)].
[30] E. Hanamura, J. Phys. Soc. Jpn. **39**, 1516 (1975).
[31] H. Akiyama, T. Kuga, M. Matsuoka, and M. Kuwata-Gonokami, Phys. Rev. B **42**, 5621 (1990).
[32] R. Leonelli, A. Manar, J. B. Grun, and B. Hönerlage, Phys. Rev. B **45**, 4141 (1992).
[33] T. Ikehara and T. Itoh, Solid State Commun. **79**, 755 (1991).

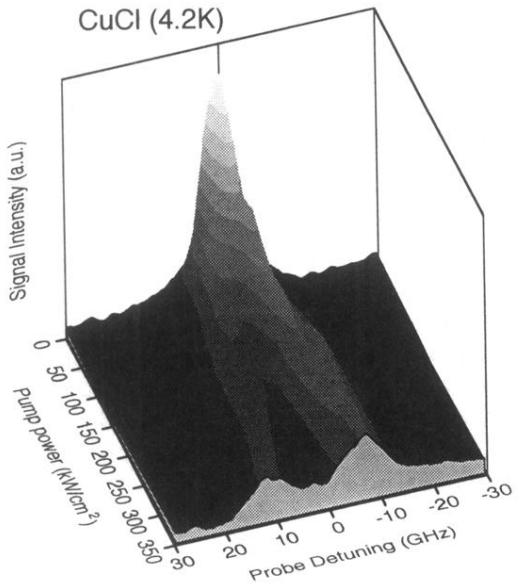

FIG. 2. TPP spectra around the biexciton line, 3.186 eV, for various pump powers. The pump detuning is almost zero.

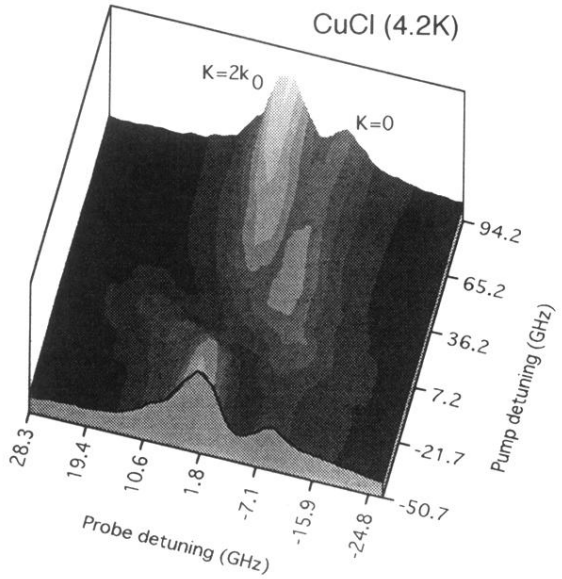

FIG. 4. Observed TPP spectra around the biexciton line for various pump laser detunings.