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Effects of exciton line widths on the amplitude of quantum beat oscillations

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In this study, we report the effects of the exciton line widths on the amplitude of quantum beat oscillations using a model with the Voigt function. Using the Voigt function, the maximum amplitude appears at the central energy between two excitons. This result agrees with many experimental results reported previously. Our analysis helps in identifying the condition for strong quantum beat oscillation, which is important for device applications such as terahertz electromagnetic wave emitters and ultrafast switches.

Exciton quantum beat is an important ultrafast coherent oscillation.^{1–15)} For instance, the frequency of these oscillations is used to decide the energy separation between two exciton states in inhomogeneous systems.¹⁶⁾ Moreover, from the device application perspective, a frequency tunable terahertz electromagnetic wave emitter^{2,9)} and an ultrafast optical switch^{8,17)} have been proposed. To generate strong quantum beat oscillations, the excitation laser is tuned to the central energy of two exciton states, which was demonstrated by detection of the terahertz wave and by a pump-probe technique.^{9,18–21)}

Given that the typical quantum beat in nanostructured semiconductors is generated by simultaneous excitation of the split heavy-hole (HH) and light-hole (LH) excitons in quantum wells, we discuss HH-LH exciton quantum beats as an example hereafter. In general, the quantum beat oscillation appearing in a pump-probe signal can be described as follows:^{1,22,23)}

$$I_{PP}(t) \propto |\mu_{HH}\mu_{LH}|^2 (1 + \cos(-\omega_{QB}t)) \exp(-\gamma_{LH-HH}t), \quad (1)$$

where μ_{HH} and μ_{LH} are the self-dipole moments of the HH and LH excitons, respectively; ω_{QB} is the quantum beat frequency corresponding to the HH-LH splitting energy; t is the time delay and γ_{LH-HH} is the dephasing rate caused by the intersubband transition between the LH and HH exciton states. Considering the dipole moment is described by the oscillator strength and the wave functions which are not explicitly a function of energy,^{24,25)} the product of $|\mu_{HH}\mu_{LH}|$ in this form does not have the peak located at the central energy between the HH and LH excitons.

On the other hand, $|\mu_{HH}\mu_{LH}|$ may also be interpreted by the spectral weight.^{1,26)} When the amplitude is given in spectral weights, the product of the laser intensities at the two

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exciton energies in the laser spectrum shows the maximum amplitude at the central energy. However, this amplitude depends on the laser intensity, which makes it difficult to explain other dependences, such as temperature dependence. Moreover, although the oscillator strength of the HH exciton is three times larger than that of the LH exciton, the maximum amplitude appears at the central energy and the number ratio of the photogenerated excitons may not determine the peak position. Therefore, in this study, we focus on the effects of the exciton line widths on the amplitude from the perspective of pump energy dependence. When the Voigt function is used for the exciton line shape, the calculated pump energy dependence agrees with the experimental results. The computed results for the quantum beat amplitude are discussed below on the basis of the exciton broadening factors.

First, the pump-energy dependence derived from various exciton waveforms is compared. Figure 1 shows the calculated results for the pump-energy dependence of the oscillation amplitude based on the Lorentzian, Gaussian, and Voigt functions. The horizontal axis is presented by the excess energy from the HH exciton. In these calculations, the HH-LH exciton splitting energy is taken to be 15 meV. The dotted lines indicate the exciton energies. In the case that the Lorentzian function is used for the HH and LH excitons, the following formula is used:

$$L_n(E) \propto \frac{1}{(E - E_n)^2 + \gamma_n^2}, \quad (2)$$

where E_n and γ_n refer to the exciton energy and the broadening factor that is associated with the homogeneous broadening, respectively. The subscript n refers to either the HH or LH excitons. The pump-energy dependence calculated as $L_{\text{HH}}L_{\text{LH}}$ indicated as the top profile in Fig. 1 shows the exciton resonance profile, with the width given by $\gamma_n = 2.0$ meV, which does not agree with the experiment results at all. Here, because the coherent overlap causing the quantum beat is given by the product of the wavefunctions,²³⁾ this product corresponds to the pump-energy dependence. This result demonstrates that the Lorentzian component determines the intensity at the exciton energy so that the ratio of the oscillator strengths hardly affects the position of the maximum amplitude at the central energy. In addition, considering that the amplitude is proportional to the product of two exciton shapes, the difference of the number of the generated excitons changes not the peak position but the maximum value.

The inhomogeneous broadening originating from the fluctuation of the well width due to the randomness in the monolayer unit is described by the Gaussian function as follows:

$$G_n(E) \propto \exp\left(-\frac{(E - E_n)^2}{2\sigma_n^2}\right), \quad (3)$$

where σ_n^2 indicates the dispersion related to the inhomogeneous broadening. While $G_{\text{HH}}G_{\text{LH}}$ in the case of $\sigma_{\text{HH}} = \sigma_{\text{LH}} = 2.5$ meV shows a peak at the central energy, the width is narrower than that in the experimental results. Therefore, although the Gaussian components contribute to the maximum at the central energy, use of the Gaussian function alone is insuf-

ficient to explain the pump-energy dependence.

Thus, to include the effects of both the Lorentzian and Gaussian components, the Voigt function is used. The Voigt function is given by the convolution of L_n and G_n and can be expressed as follows:

$$V_n(E) \propto \int L_n(E)G_n(E - E')dE'. \quad (4)$$

The explicit form of the function used for the calculation^{27,28)} is shown as follows:

$$V_n(E) \propto \frac{\text{Erf}\left(\frac{-[(E-E_n)+E']}{\sqrt{2}\sigma_n}\right)}{2[(E-E_n)^2 + \gamma_n^2]} - \frac{\text{Erf}\left(\frac{-[(E-E_n)-E']}{\sqrt{2}\sigma_n}\right)}{2[(E-E_n)^2 + \gamma_n^2]}. \quad (5)$$

The $V_{\text{HH}}V_{\text{LH}}$ product indicated at the bottom of Fig. 1 shows a peak with moderate width at the center between the exciton energies. Therefore, comparing the experimental result plotted by the open circles,¹⁸⁾ the Voigt function is a reasonable choice to discuss the effects of the exciton line width.

Here the role of the Gaussian component is discussed. Figure 2(a) shows the calculated results for various σ values and a constant γ_n value of 2.0 meV. With an increasing σ value, $V_{\text{HH}}V_{\text{LH}}$ increases, and excitonic peaks appear in the profiles of $\sigma \geq 4.0$ meV. To clarify the dependence on σ , the value of $V_{\text{HH}}V_{\text{LH}}$ is plotted in Fig. 2(b) as a function of σ_n . The solid and dotted curves indicate the values at the central and exciton energies, respectively. When $\sigma_n \leq \gamma_n$, both values of $V_{\text{HH}}V_{\text{LH}}$ increase gradually. On the other hand, in the case of $\sigma_n > \gamma_n$, the amplitudes at the exciton energies increase rapidly in comparison with the amplitude at the central energy. Therefore, to generate the intense quantum beat, larger σ values may be better, because it is possible to excite the larger numbers of excitons within inhomogeneous widths. Moreover, the increase in σ corresponds to that in the generated excitons so that the amplitude of the quantum beat depends on the generated exciton number. As a result, when the number of generated HH excitons is different from that of LH excitons, the smaller number decides the amplitude. This is the reason for the insensitivity of the peak energy to the ratio of the oscillator strength.

Next, the role of the Lorentzian component is discussed. Figure 3(a) demonstrates the pump energy dependence of the quantum beat amplitude for a constant σ_n value of 2.5 meV and various γ values. With increasing γ_n , the peaks at the exciton energies disappear, whereas that at the central energy prevails; the resulting profile changes from the exciton resonance characteristic to a single peak. The $V_{\text{HH}}V_{\text{LH}}$ values at the central and the exciton energies are plotted as a function of γ_n in Fig. 3(b), represented by solid and dotted curves, respectively. With increasing γ_n , the amplitude at the central energy decreases gradually, whereas that at the exciton energies decreases rapidly. Therefore, unless γ_n is less than half of σ_n , which is difficult in samples with high purity and small thickness fluctuations at the interfaces, suppression of the inhomogeneous width in the growth process hardly contributes

to the generation of strong oscillations. In addition, because generation of the quantum beat increases the value of γ due to the intersubband transition,²⁸⁾ a reduction in γ for strong oscillations is not easy to achieve. Although the values of γ_{HH} and γ_{LH} are usually different, same value was used for the calculation. When the γ value increases, the Lorentzian shape spreads and the peak value decreases. Hence, in the case of that the values are the same order, the difference of the γ value changes only the intensity at the exciton energies and hardly changes the profile.

In summary, we have investigated the effects of exciton line shapes on the amplitude of quantum beat oscillation. The Voigt function, including the inhomogeneous and homogeneous broadening factors, could demonstrate results similar to the experimental results, which showed a peak at the central energy between two excitons. Moreover, to enhance the oscillation amplitude, the inhomogeneous width was found to be an important factor, because the number of photogenerated excitons increases. Our results help in the determination of a valid value for the exciton widths in order to generate strong oscillations.

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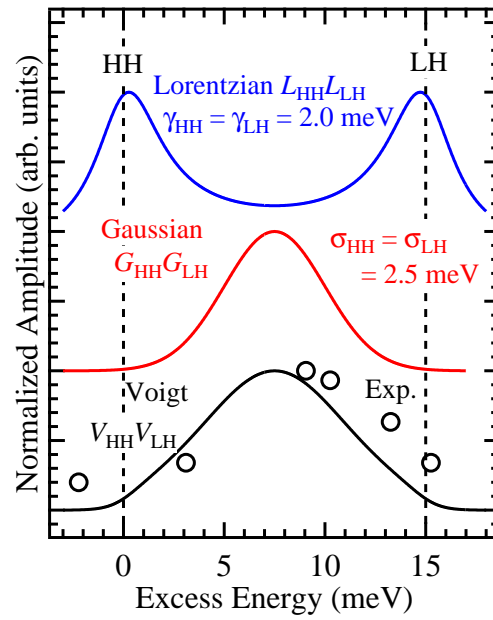


Fig. 1. Dependence of the calculated quantum beat amplitude on the pump energy. The horizontal axis is represented by the excess energy from the HH exciton. The dotted lines indicate the exciton energies. From the top, the Lorentzian, Gaussian, and Voigt functions are used to represent the exciton line profiles. The open circles indicate the experimental result reported in Ref. [18].

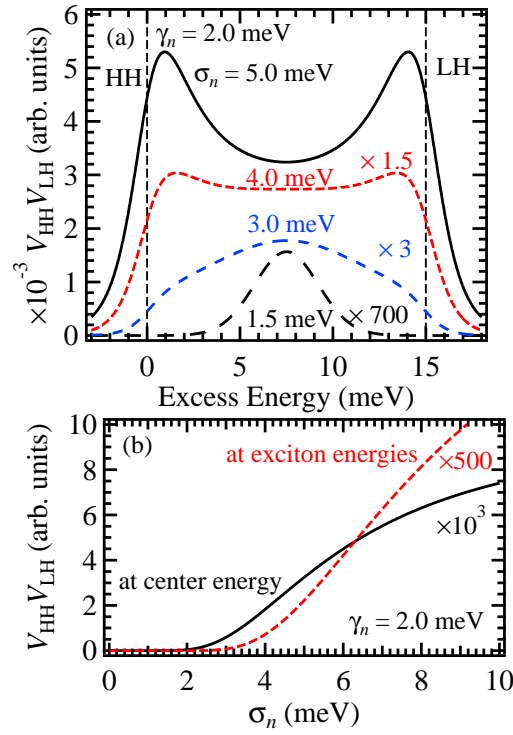


Fig. 2. (a) Dependence of the calculated amplitude on the pump energy using $\gamma_n = 2.0$ meV and various σ_n values. The vertical dotted lines indicate the exciton energies. (b) The amplitudes at the central and exciton energies are plotted as a function of σ_n , represented by solid and dotted curves, respectively.

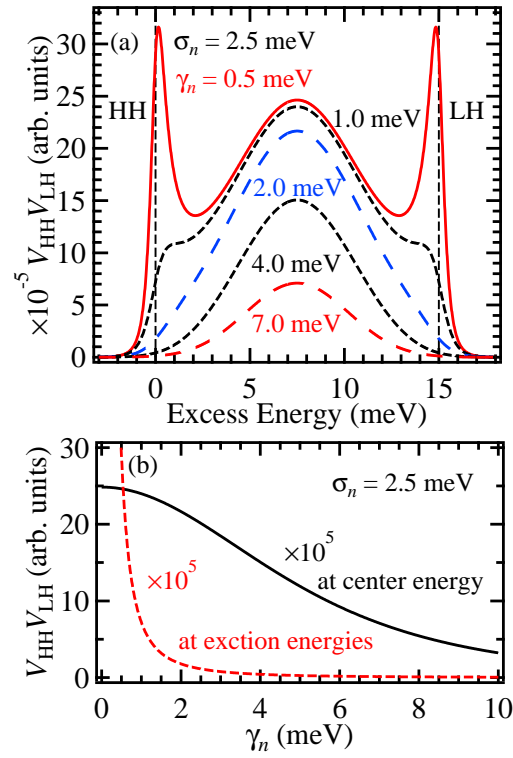


Fig. 3. (a) Dependence of the calculated amplitude on the pump energy using $\sigma_n = 2.5$ meV and various γ_n values. The vertical dotted lines indicate the exciton energies. (b) The amplitudes at the central and exciton energies are plotted as a function of γ_n , represented by the solid and dotted curves, respectively.

References

- 1) K. Leo, E. O. Göbel, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. F. Müller, K. Köhler, and P. Ganser, *Phys. Rev. B* **44**, 5726 (1991).
- 2) P. C. M. Planken, M. C. Nuss, I. Brener, K. W. Goossen, M. S. C. Luo, S. L. Chuang, and L. Pfeiffer, *Phys. Rev. Lett.* **69**, 3800 (1992).
- 3) I. Brener, P. C. M. Planken, M. C. Nuss, L. Pfeiffer, D. E. Leaird, and A. M. Weiner, *Appl. Phys. Lett.* **63**, 2213 (1993).
- 4) E. J. Mayer, G. O. Smith, V. Heukeroth, J. Kuhl, K. Bott, A. Schulze, T. Meier, S. W. Koch, P. Thomas, R. Hey, and K. Ploog, *Phys. Rev. B* **51**, 10909 (1995).
- 5) T. Dekorsy, A. M. T. Kim, G. C. Cho, S. Hunsche, H. J. Bakker, H. Kurz, S. L. Chuang, and K. Köhler, *Phys. Rev. Lett.* **77**, 3045 (1996).
- 6) A. V. Gopal and A. S. Vengurlekar, *Phys. Rev. B* **62**, 4624 (2000).
- 7) A. G. V. Spivey and S. T. Cundiff, *J. Opt. Soc. Am. B* **24**, 664 (2007).
- 8) O. Kojima, T. Isu, J. Ishi-Hayase, A. Kanno, R. Katouf, M. Sasaki, and M. Tsuchiya, *J. Phys. Soc. Jpn.* **77**, 044701 (2008).
- 9) S. Priyadarshi, K. Pierz, U. Siegner, P. Dawson, and M. Bieler, *Phys. Rev. B* **83**, 121307 (2011).
- 10) T. Hasegawa, Y. Takagi, and M. Nakayama, *Phys. Rev. B* **83**, 205309 (2011).
- 11) S. Ohta, O. Kojima, T. Kita, and T. Isu, *J. Appl. Phys.* **111**, 023505 (2012).
- 12) O. Kojima, K. Mizoguchi, and M. Nakayama, *J. Appl. Phys.* **112**, 043522 (2012).
- 13) S. Khan, J. Jayabalan, R. Chari, S. Pal, S. Porwal, T. Kumar S. and S. M. Oak, *Appl. Phys. Lett.* **105**, 073106 (2014).
- 14) D. Shyshlov, E. Berrois, M. Gruebele, and D. Babikov, *J. Chem. Phys.* **141**, 224306 (2014).
- 15) N. Thakkar, C. Cherqui, and D. J. Masiello, *ACS Photonics* **2**, 157 (2014).
- 16) T. Lai, L. Teng, Z. Jiao, H. Xu, L. Lei, J. Wen, and W. Lin, *Appl. Phys. Lett.* **91**, 062110 (2007).
- 17) O. Kojima, K. Hayashii, T. Kita, and K. Akahane, *J. Phys. D: Appl. Phys.* **47**, 105101 (2014).
- 18) O. Kojima, K. Mizoguchi, and M. Nakayama, *Phys. Rev. B* **68**, 155325 (2003).
- 19) O. Kojima, K. Mizoguchi, and M. Nakayama, *J. Lumin.* **108**, 195 (2004).
- 20) O. Kojima, K. Mizoguchi, and M. Nakayama, *Phys. Rev. B* **70**, 233306 (2004).
- 21) O. Kojima, K. Mizoguchi, and M. Nakayama, *J. Lumin.* **112**, 80 (2005).

- 22) K. Leo, J. Shah, T. C. Damen, A. Schulze, T. Meier, S. Schmitt-Rink, P. Thomas, E. O. Göbel, S. L. Chuang, M. S. C. Luo, W. Schäfer, K. Köhler, and P. Ganser, IEEE J. Quantum Electron. **28**, 2498 (1992).
- 23) J. Shah, Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures, 2nd ed., edited by M. Cardona, Springer Series in Solid-State Sciences, Vol. 115 (Springer-Verlag, Berlin, 1999), Chap. 2.
- 24) Y. Masumoto, M. Matuura, S. Tarucha, and H. Okamoto, Phys. Rev. B **32**, 4275 (1985).
- 25) J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moor, C. Foxon, and R. J. Elliot, Phys. Rev. Lett. **59**, 2337 (1987).
- 26) T. Hasegawa, K. Mizoguchi, and M. Nakayama, Phys. Rev. B **76**, 115323 (2007).
- 27) A. D. Bristow, D. Karauskaj, X. Dai, R. P. Mirin, and S. T. Cundiff, Phys. Rev. B **79**, 161305 (2009).
- 28) O. Kojima, K. Kojima, T. Kita, and K. Akahane, Phys. Rev. B **91**, 125307 (2015).