Photomodulation spectroscopy for determining the total exciton absorption in single quantum wells

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The photomodulation spectra of single quantum wells based on InGaAs/GaAs have been studied for pump power densities from 0.5 to 300 mW/cm². It is found that the spectra measured at different pump powers differ not only in amplitude, but also in spectral position. Moreover, the dependence between the amplitude and the spectral position of the modulation spectrum is linear. We explain this dependence in terms of the generally accepted model of the modulation spectra, with allowance for the second-order expansion of the exciton contribution to the absorption and the refraction over the exciton resonance frequency. By constructing the dependence of the spectral position of the modulation spectrum as a function of its amplitude, we can find the limiting position of the exciton resonance as the amplitude tends toward zero. This limit corresponds to the spectral position of the exciton resonance in the absence of pump radiation. We found that the observed spectral shift relative to the position in the absence of pumping equals half the shift of the exciton resonance frequency caused by external modulation. Using the modulation of the exciton resonance frequency determined in this manner, the calculated values of the derivatives \( \partial R/\partial n \) and \( \partial R/\partial \delta k \), and the modulation spectrum \( \Delta R/R \), it is easy to determine the absolute value of the exciton absorption (the oscillator strength of the exciton) in a quantum well. © 1996 American Institute of Physics. [S1063-7826(96)00601-9]

Modulation spectroscopy is widely used to study structures with single quantum wells (QWs) (see the review in Ref. 1 and the references cited there). This technique possesses high sensitivity and makes it easy to record the spectra of single QWs at room temperature. The conventional means of processing the modulation spectra of QWs makes it possible to determine the spectral position, the width, and the shape of the exciton absorption peak. It is usually assumed that the total absorption of these peaks cannot be determined from the modulation spectra, because the amplitude of the spectra is proportional not only to the amplitude of the exciton absorption peak, but also to the shift of the exciton resonance frequency caused by the modulation. Since the latter value is generally unknown, it is impossible to determine the total absorption from these spectra.

We have found that for the photomodulation spectra of structures with single InGaAs/GaAs quantum wells, varying the pump power density from 0.5 to 300 mW/cm² not only increases the amplitude of the spectra, but also increases the frequency shift of the spectrum as a whole. In this case, there is a linear dependence between the spectral position of the modulation spectrum and its amplitude (Figs. 1 and 2). The parameters of the samples are shown in Table I, and the experimental setup is described in Refs. 2 and 3.

Under the conditions of the experiment, the QW is located in the region of the surface space charge, the pump power density does not exceed 0.3 W/cm², and the photoreflection spectrum is formed mainly by modulating the exciton resonance frequency and to a slight extent by modulating the other exciton parameters—the spectral width and the total absorption in the exciton peak. The technique proposed below for processing the modulation spectra depends only on this circumstance, is rather general, and is not associated with a specific modulation mechanism. As a result, this technique can be used, on the basis of the modulation measurements, to determine not only the position and width of the exciton resonances, but also the total absorption in the exciton peak in structures with single QWs. Such a broadening of the possibilities of modulation spectroscopy, in our view, is crucial. For example, this technique can be used to study structures with strongly directional layers, when it is difficult to grow a structure containing the large number of layers needed to directly measure absorption in a QW.

To explain the observed correlation between the amplitude and the spectral position of the modulation spectra, we use the conventional model for the photoreflection spectra, taking into account the second-order expansion of the frequency dependence of the optical constants of the QW in the exciton resonance frequency. The contribution of the exciton peak to the contours of the absorption \( k(\omega) \) and the refractive index \( n(\omega) \) can be written as

\[
k(\omega) = S \frac{1}{\pi} \frac{\Gamma}{(\omega_{ex} - \omega)^2 + \Gamma^2};
\]

\[
n(\omega) = S \frac{1}{\pi} \frac{\omega_{ex} - \omega}{(\omega_{ex} - \omega)^2 + \Gamma^2},
\]

where \( S = \int k(\omega) d\omega \), \( \omega_{ex} \), and \( \Gamma \) are the resonance frequency and the phenomenological damping of the exciton. Here, the exciton absorption peak has a Lorentzian shape. We should point out that the proposed treatment can be extended to the Gaussian case or any other absorption line contour with no difficulty.
The total absorption in the exciton peak in a well of width \( L \) is
\[
S_0 = \frac{4\pi \omega_{\text{ex}} L}{\lambda_{\text{ex}}} = \frac{2\omega_{\text{ex}}}{c} L S,
\]
where \( \lambda_{\text{ex}} \) is the wavelength of light in vacuum, corresponding to the frequency \( \omega_{\text{ex}} \).

The normalized spectral variation of the reflection can be represented as a linear combination of the spectral variation of the refractive index and the absorption coefficient:
\[
\Delta R(\omega) = \frac{R_1(\omega) - R_0(\omega)}{R_0(\omega)} = \alpha \Delta n(\omega) + \beta \Delta k(\omega),
\]
where \( R_0(\omega) \) is the spectral reflectance without pumping and \( R_1(\omega) \) is the spectral reflectance with pumping. The coefficients \( \alpha \) and \( \beta \) can be obtained by considering the reflection from a multilayer structure which consists of a covering...
layer with a QW and a substrate. In computing the reflection of light from the multilayer structure and the derivatives \( \partial R/\partial n \) and \( \partial R/\partial k \), we assumed that the QW can be represented as a homogeneous layer of thickness \( L \) with a certain effective refractive index. A rigorous computation of the reflection of light from a QW shows that such a representation is acceptable when the angles of incidence are close to normal.

Representing the functions in Eq. (1) as expansions in Taylor series up to the second order in \( \omega_{ex} \), we find \( \Delta n(\omega) \) and \( \Delta k(\omega) \)

\[
\Delta n(\omega) = \frac{\partial n}{\partial \omega_{ex}} \Delta \omega_{ex} + \frac{1}{2} \frac{\partial^2 n}{\partial \omega_{ex}^2} \Delta \omega_{ex}^2
\]

\[
\approx \frac{\partial}{\partial \omega_{ex}} \left[ n \left( \omega + \frac{1}{2} \Delta \omega_{ex} \right) \right] \Delta \omega_{ex},
\]

\[
\Delta k(\omega) = \frac{\partial k}{\partial \omega_{ex}} \Delta \omega_{ex} + \frac{1}{2} \frac{\partial^2 k}{\partial \omega_{ex}^2} \Delta \omega_{ex}^2
\]

\[
\approx \frac{\partial}{\partial \omega_{ex}} \left[ k \left( \omega + \frac{1}{2} \Delta \omega_{ex} \right) \right] \Delta \omega_{ex}.
\]

As \( \Delta \omega_{ex} \) increases, the modulation spectrum of the reflection,

\[
\Delta R(\omega) = \left[ \alpha \frac{\partial}{\partial \omega_{ex}} \left[ n \left( \omega + \frac{1}{2} \Delta \omega_{ex} \right) \right] \right] \Delta \omega_{ex}
\]

\[
+ \beta \frac{\partial}{\partial \omega_{ex}} \left[ k \left( \omega + \frac{1}{2} \Delta \omega_{ex} \right) \right] \Delta \omega_{ex}
\]

increases in amplitude in proportion to \( \Delta \omega_{ex} \) and shifts as a whole by \( 1/2 \Delta \omega_{ex} \). It can be seen that such a shift is not explained when it is expanded to the first order (which is traditionally used for processing modulation spectra). Such a correlation between the amplitude and the spectral position of the modulation spectrum makes it possible to experimentally determine the shift of the exciton peak caused by external modulation.

The procedure for determining \( \Delta \omega_{ex} \) and the total absorption in the exciton peak can be described as follows. For each of the spectra measured at different pump powers, we use a curve-fitting method to find the amplitude \( A \) and the spectral position \( \omega_f \). Having a set of \( A \) and \( \omega_f \) parameters for all the measured spectra and knowing that \( A \) and \( \omega_f \) are associated by a linear dependence because of Eq. (6), we find the limiting value of \( \omega_{ex} \) as \( A \rightarrow 0 \). The \( \omega_{ex} \) thus determined corresponds to the position of the exciton resonance \( \omega_{ex0} \) in the absence of pumping. The displacement of the exciton resonance due to pumping is \( \Delta \omega_{ex} = 2(\omega_f - \omega_{ex0}) \). If \( \Delta \omega_{ex} \) is known, it is possible, starting from Eqs. (6), (4), and (1), to compute the total absorption in the exciton peak.

The results of processing the spectra are shown in Table I. Sample Z1371 differs from sample Z1022 by the presence of a \( \delta \)-doped \( n \)-type layer 1000 \( \AA \) from the surface of the sample. This layer was introduced into the structure in order to increase the built-in field. The reduction of the total absorption in the exciton peak can therefore probably be explained in terms of screening of the exciton by free electrons.

We should point out that, according to theoretical estimates, the total absorption in the exciton peak depends upon the size and binding energy of the excitons;\(^7\) \( \ldots \) the experimentally measured total absorption in the exciton peak can be used to determine the exciton parameters. For the InGaAs/GaAs QWs being investigated, the exciton binding energy amounts to 7–8 meV (Ref. 9), and the total absorption in the exciton peak is about 0.2 meV.

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