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OPTICAL GAIN/ABSORPTION AND FANO INTERFERENCE IN HIGH DENSITY EXCITON SYSTEM IN CuCl

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Optical gain/absorption measurements in CuCl at 1.7K show an increasing dissociation of biexcitons into free excitons with increasing excitation density and an apparent saturation of the total density of e-h pairs in the exciton-biexciton system. At high excitation density, a Fano interference is observed between a discrete bound exciton line and a non-linear continuum background.

1. Introduction

The high density exciton - biexciton system in CuCl has in recent years been studied extensively by luminescence, two-photon absorption and two-photon Raman scattering [1]. The very nature of a high density exciton system strongly coupled to photons (polaritons) implies that spectral line shapes and kinetics may be strongly influenced by stimulation of the transitions. Thus, stimulated emission [2] as well as induced absorption [3] have been reported from exciton - biexciton transitions (M band) in CuCl. In the present work, we investigate in detail the optical gain/absorption in the spectral range of bound exciton and biexciton emission.

2. Experiment

The absorption/gain spectra were obtained by an alternating-excitation-length technique previously described [4]. The excitation source was a pulsed N_2-gas laser (100 kW, 10 ns, 100 Hz). The excitation length was about 0.2 mm and the maximum excitation density J = 10 MW/cm^2. The crystals used in the experiments were high-quality platelets grown by vapour-phase transport of ultra-pure CuCl. They were immersed in liquid helium pumped to 1.7K.

3. Results

Figure 1 shows absorption/gain spectra obtained at different excitation densities ranging from 10 kW/cm^2 to 10 MW/cm^2. For comparison is also shown a luminescence spectrum as emitted from the excited surface (100 kW/cm^2) of the sample. In Fig. 1, several pronounced spectral features show up in luminescence as well as in absorption/gain. Well known structures associated with bound exciton transitions (ν_2 [5] and ν_3 [6]) and exciton - biexciton transitions (M_T and M_L [7]) are indicated by arrows in the figure. The luminescence structures at 3.18 - 3.19 eV and 3.14 - 3.15 eV are, however, not well understood. The width of the structures and their kinetics (non-saturating) suggest intrinsic processes for the emission in these bands.

The pronounced structures in the absorption/gain spectra are very sensitive to changes in the excitation power. They appear on a continuum background of absorption and gain which is not previously reported and the origin of which is not clearly understood. In order to analyze the line shape and kinetics of the different elements in the absorption/gain spectra, they have to be separated out from the background. This is not easily done, because negative as well as positive contributions to the spectra are present. However, the separation of the spectra into impurity lines, M band and a continuum background is rather obvious in the low excitation case, and the same elements are then traced in the spectra for increasing excitation density. The results are shown in Fig. 2.

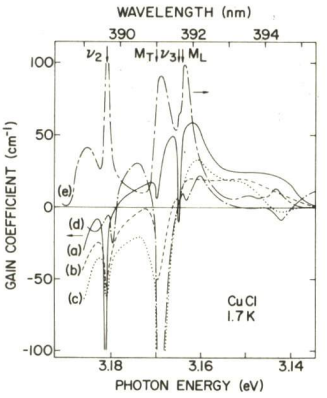

Figure 1

Absorption/gain spectra for different excitation powers (a): 10kW/cm^2, (b): 100kW/cm^2, (c): 1MW/cm^2, (d): 10MW/cm^2, and a luminescence spectrum (e) recorded at 100kW/cm^2

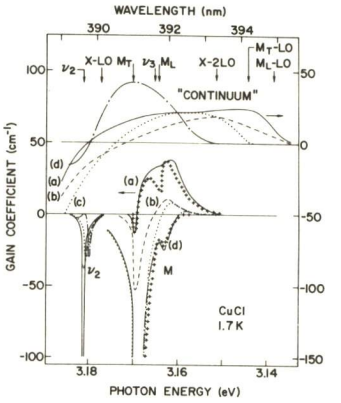

Figure 2

Separate absorption/gain spectra for M band, ν_2 line and background continuum. The crosses are spectra calculated from eq. (1) with $E_M - E_X(\Gamma_{5T}) = 3.170$eV, $\Delta_{LT} = 6.3$eV and (a): $g_0 = 0.345$ eV cm^{-1}, $\eta = 0.5$, $\beta = 0.39$ meV^{-1}; (b): $g_0 = 0.045$ eV cm^{-1}, $\eta = 16$, $\beta = 0.31$ meV^{-1}

In the M band, we observe positive gain at low excitation density which, with increasing excitation density, is progressively quenched from the high energy side and converted into induced absorption. The ν_3 absorption line, which has been removed in Fig. 2, diminishes with increasing excitation probably due to saturation of the absorbing centres. In contrast to that, the integrated intensity of the ν_2 line stays fairly constant. It is, however, broadened strongly with increasing excitation until it abruptly changes shape from a symmetric line around the resonance into a strongly asymmetric line shape featuring a transmission peak as well as an absorption peak near the resonance.

4. Discussion

The M band spectra in Fig. 2 can be compared with a simple theory neglecting polariton effects and assuming Boltzmann distribution of excitons and biexcitons, within and between their respective bands, at an effective temperature T_x [8,9]. Including the longitudinal-transverse (LT) splitting $\Delta_{LT}=E_x(\Gamma_{5L})-E_x(\Gamma_{5T})$ of the Γ_5 free excitons, theory gives:

$$g(\hbar\omega) = g_o\beta^{3/2}(1-2^{3/2}\eta e^{-\beta E})E^{1/2}L(E)e^{-\beta E} \quad , \tag{1}$$

where $E = E_M - E_x(\Gamma_{5T}) - \hbar\omega > 0$, E_M is the biexciton ground state energy and $\beta = 1/kT_x$. The parameter $\eta = N_x(\Gamma_{5T})/N_M$ is the ratio between the total densities of Γ_{5T} excitons and biexcitons. The function L(E) accounts for the LT splitting of the M band and is unity for $0<E<\Delta_{LT}$. For $E > \Delta_{LT}$, $L(E) = 1 + 0.5\ (1 - \Delta_{LT}/E)^{1/2}e^{\beta\Delta}{}_{LT}$. The prefactor in eq. (1) contains N_M as well as the oscillator strength f_M (assumed wave vector independent [10]) for the exciton - biexciton transitions:

$$g_o = (2e^2\pi^{1/2}h/m_o nc)\cdot N_M \cdot f_M \tag{2}$$

where e and m_o are charge and rest mass of the electron and n is the refractive index of the material. Gain spectra calculated according to eq. (1) are shown in Fig. 2 (crosses) for two sets of parameters β, η and g_o selected by visual fitting to the experiment for low (a) and high (d) excitation density. The basis for our fit are the well known ground state energies $E_x(\Gamma_{5T}) = 3.2025$ eV, $E_x(\Gamma_{5L}) = 3.2080$ eV and $E_M = 6.3720$ eV [1,3,7]. Probably due to polariton effects, an improved fit was obtained by increasing slightly the LT splitting to $\Delta_{LT} = 6.3$ meV. Despite the simplicity of the model, it explains the gross features of the observed spectra quite well. From a number of such fits, we have determined the values of the parameters β, η, and g_o and their dependences on excitation power. The results are shown in Fig. 3. From the value $g_o = 0.345$ eVcm^{-1} at J = 10 kW/cm^2, we find from eq. (2) $N_M \cdot f_M = 6.5 \times 10^{15}$ cm^{-3} yielding $f_M \simeq 1$ in agreement with previous reports [3]. In this estimate we have assumed life time $\tau = 100$ ps and diffusion length 1 μm in the exciton - biexciton system.

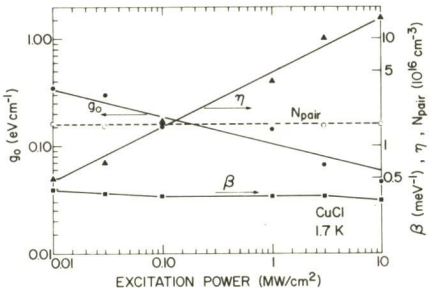

Figure 3

Exciton - biexciton parameters from fitting the line shape eq. (1) to experiment at different excitation powers: Lines are drawn to guide the eye

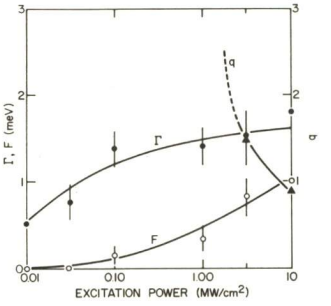

Figure 4

Parameters of Fano line shape eq.(3) fitted to the ν_2 line for different excitation powers: The error bars are estimated and the curves to guide the eye only

The variation of η and g_o in Fig. 3 shows that the density of Γ_5 excitons is only increased by a factor of 4.2 and the density of biexcitons is even diminished by a factor of 7.7 when the excitation density is increased over three orders of magnitude. A similar behaviour has previously been observed in ZnO [11], but it is in obvious contradiction to a simple kinetics model. It is interesting to note that the density $N_{pair} = N_M(2 + \eta(1 + 0.5\exp\ -\beta\Delta_{LT}\ ))$ of e-h pairs in the Γ_5 exciton - biexciton system seems to be constant in our experiment. Taking $f_M = 1$ as above, we have calculated N_{pair} for a series of spectral fits with the result $N_{pair} \approx 1.6 \times 10^{16}$cm^{-3}, independently of excitation power as shown by the open circles in Fig. 3. At present it is not clear whether the observed

constancy of N_{pair} is a real pinning effect or just the result of an accidental balance between transitions which then have to include processes other than those already considered. Such processes are inelastic exciton - exciton, excition - electron and possibly also biexciton collisions. These processes may in turn contribute to the observed absorption/gain continuum.

The ν_2 line broadening, shift and final asymmetric distortion, which occurs simultaneously with the growing of a continuum background absorption in the same spectral region, points towards a Fano interference [12] between the continuum background and the discrete level. We have compared the observed ν_2 line shapes with a Fano-type line shape of the form [12]:

$$\alpha_I \propto (q + \varepsilon)^2/(1 + \varepsilon^2) \quad , \tag{3}$$

where $\varepsilon = (\hbar\omega - E_r)/0.5\Gamma$ is the energy deviation from the resonance at E_r normalized to half the width of the autoionized level. The resonance E_r is shifted with respect to the independent resonance $E(\nu_2)$ by an amount F, i.e. $E_r = E(\nu_2) - F$. The parameter q expresses the strength of the transition probability to the modified discrete level relative to a band width Γ of the continuum. The comparison gives parameters Γ, F and q that are dependent on excitation density as shown in Fig. 4. We therefore suggest that the interference is with a two-polariton continuum [13] which indeed gives rise to a non-linear absorption in the spectral range of the ν_2 line. In fact, absorptive two-polariton transitions are strongly enhanced at photon energies $\hbar\omega = E_M/2$ (resonant two-photon absorption) and also around $\hbar\omega = E(M_T) = E_M - E_x(\Gamma_{5T})$ (induced absorption), i.e. on both sides of the ν_2 line.

5. Summary

The optical gain and induced absorption observed in the M band of CuCl is consistent with well established values for biexciton energy $E_M = 6.3720$ eV, effective temperature $T_x = 30$ - 35 K and oscillator strength $f_M = 1$. The kinetics, however, indicate a saturation of the total pair density in the exciton - biexciton system at $N_{pair} = 1.6 \times 10^{16}$ cm^{-3} which is not explained so far. This saturation may be related to the observed interference between the bound exciton line ν_2 and a continuum background.

Footnotes and References

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