Journal of Advances in Physics Vol 18 (2020) ISSN: 2347-3487 https://rajpub.com/index.php/jap

DOI: https://doi.org/10.24297/jap.v18i.8805

Extraction of temperature-dependent exciton-polariton damping in InP bulk crystal

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Abstract

The temperature dependence of exciton-polariton damping in InP bulk crystal was extracted by the method of integrated absorption. The extraction procedure excluding the contribution of inhomogeneous broadening into the exciton ground state absorption linewidth is graphically illustrated. The extracted temperature-dependent damping is analyzed regarding the primary dissipative mechanism in order to determine the material parameters of exciton-polariton scattering by acoustic and optical phonons.

Keywords: exciton-polaritons, integrated absorption, Indium phosphide

Introduction

Indium phosphide (InP) semiconductor material is successfully adopted in the manufacturing of high-power and high-speed electronic products Gao (2015). Numerous optoelectronic and photonic devices based on InP and its heterostructure systems have been developed and are widely used today Kasap (2017). Particular optical properties of InP compound makes it promising Sanvitto (2016) for future exciton-polariton device applications utilizing quasiparticle combination of light and matter.

Even though the extensive research for the last two decades intended to eliminate a lack of definite knowledge of many material parameters of III-V semiconductor compounds Adachi (1992), the experimental verification of exciton-polariton parameters by optical methods remains relevant nowadays.

The experimental study of exciton resonances face a problem of inhomogenous broadening of exciton lines due to the physical and crystal imperfection of the fabricated samples of a semiconductor material. Inhomogenous broadening prevails over the true homogenous broadening contributed by dissipative mechanisms of exciton-polaritons and strongly hampers determination of damping parameters.

The given work demonstrates the procedure for extraction of the temperature dependence of exciton-polariton damping excluding the contribution of inhomogeneous broadening from the observed exciton absorption line in InP bulk semiconductor material by the method of integrated absorption. The method was empowered by theoretical consideration of the role of spatial dispersion in the absorption of light by excitons Akhmediev (1980) and developed studying the exciton-polariton light transfer in a variety of semiconductor bulk crystals Seisyan (2020), Gorban' (2000), solid solutions Seisyan (2005) and heterostructures with multiple quantum wells Kosobukin (1993), Vaganov (2011).

The extracted damping is analyzed versus temperature regarding main dissipative mechanisms in order to obtain material parameters of exciton-polaritons in InP bulk crystals.

Experimental

The temperature dependent integrated absorption was studied in InP semiconductor samples of high purity and perfection. The bulk crystals were grown by vapor phase epitaxy. The free standing thin InP layers were separated from substrates by a wet chemical etching. Finally, the samples were annealed in hydrogen and kept freely packed in a box made of cover glass. Using this fabrication technique it is possible to obtain ultra thin samples free of inhomogeneous deformations and residual stress. Inhomogeneous broadening of spectral line is inherent to any resonant excitation localized over the ensemble of particles with distinct local parameters and conditions. In bulk semiconductors the main inhomogeneities are deformations, local atomic environment, charge state-distributions, and electric field of surface charge. It likely to significantly reduce inhomogenous broadening in semiconductors using advanced fabrication technologies but not to eliminate it completely.

The optical layout and experimental setup is described in Vaganov (2011). The exciton absorption α(ω) was studied in the temperature range from 5K to 300 K. The ground state exciton line was separated from the total spectrum of the fundamental absorption edge by the methods of contour analysis Markosov (2009) and integrated numerically.

Integrated Absorption of Exciton-Polaritons

According to the phenomenological model Akhmediev (1980) the propagation of light through semiconductor crystals is characterized by exciton-polariton nature and described by two-wave mode due to the spatial dispersion induced by nonzero exciton translational mass Pekar (1958). The integrated absorption $K(\Gamma)$ increases with damping Γ until the critical damping Γ_c is reached, after which $\Gamma > \Gamma_c$ the light transfer is actually performed only by single wave with dispersion independent on wavevector, so that integrated absorption becomes constant $K(\Gamma > \Gamma_c) = K_{max}$ Seisyan (2020).

The experimental dependence of integrated absorption $K(T)$ is shown at Fig. 1 by blue dots with a line connecting experimental points. The dependence becomes saturated $K(T > T_c) = K_{max}$ above the critical temperature T_c. The theoretical dependence of integrated absorption $K(\Gamma)$ calculated with the experimentally obtained value of $K_{max} = K(T > T_c) = K(\Gamma > \Gamma_c)$ is shown in Fig. 1 by a green line.

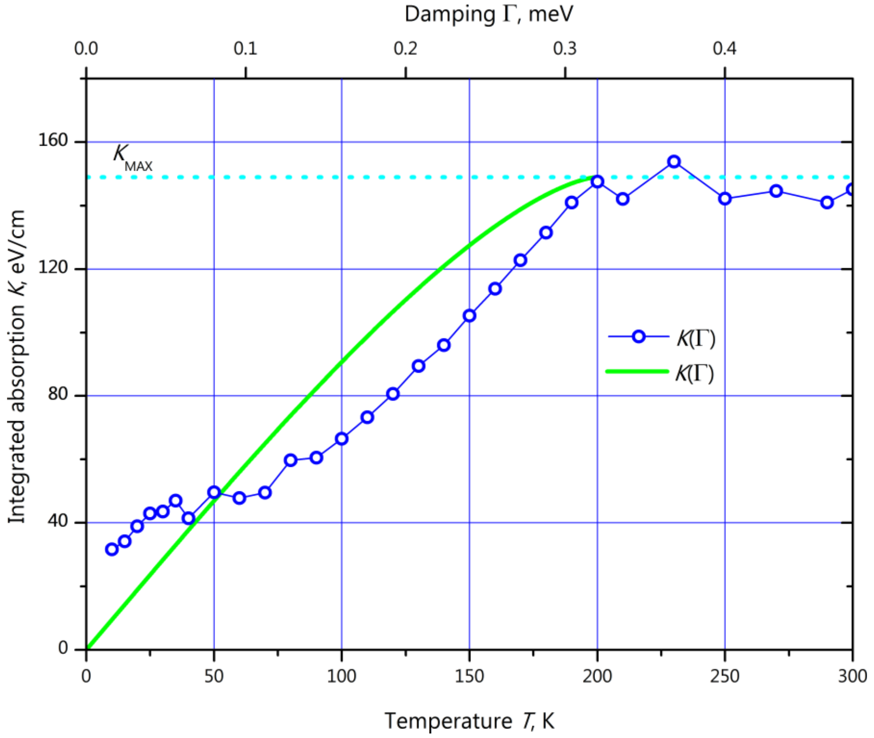

Figure 1. The experimental dependence of integrated absorption $K(T)$ is shown by blue dots with a line connecting experimental points. The theoretical dependence of integrated absorption $K(\Gamma)$ Calculated with the experimentally obtained value of $K_{max} = 149$ eV/cm and $\Gamma_c = 0.32$ is shown by a green line. The maximum value of integrated absorption K_{max} is designated by a dashed cyan line.

The critical damping Γ_c and the longitudinal transverse splitting $\hbar\omega_{LT}$ was derived Akhmediev (1980) from the maximum value of experimentally obtained integrated absorption $K_{max} = K(T > T_c)$ as

$$\Gamma_c = 2\hbar\omega_0\sqrt{2\varepsilon_b\frac{\hbar\omega_{LT}}{Mc^2}} \quad , \quad \hbar\omega_{LT} = K_{\max}\frac{c\hbar}{\pi\sqrt{\varepsilon_b}\cdot\hbar\omega_0} \qquad (1),$$

where $\hbar\omega_0$ is the energy of exciton resonance, ε_b is the background dielectric constant, c is the speed of light, and M is the exciton translational mass. Both the values of critical damping Γ_c and the longitudinal-transverse splitting $\hbar\omega_{LT}$ are determined with high precision as experimentally obtained maximum value of integrated absorption K_{max} is taken from the saturation region $T > T_c$.

Extraction Procedure and Material Parameters of Exciton-Polaritons in InP

Due to the inhomogenous broadening, the observed linewidth of exciton ground state absorption line $H(T_c)$ at the critical temperature T_c is by an order of magnitude more than the critical damping Γ_c .

The inhomogenously broadened lineshape in semiconductor materials is often represented as a convolution of a homogenously broadened line with a function of Gaussian or more complex distribution following the implementation of the random processes with the major contribution into the observed linewidth. But actually the deconvolution to the true line could be reliably performed only for special cases and require a prior knowledge about the original lineshape and the parameters of the broadening function.

The light transfer above the critical damping $\Gamma>\Gamma_c$ is performed by single wave, so excitonic absorption line has well known Lorentzian shape. In exciton-polariton mode below the critical damping $\Gamma<\Gamma_c$ the lineshape is complex and is difficult to be expressed analytically Akhmediev (1980).

The method of integrated absorption enables direct determination of the temperature dependence of damping $\Gamma=\Gamma(T)$ instead of reducing to the true homogenously broadened spectral lineshape and analyzing the dependence of its linewidth on temperature.

The extraction procedure of exciton-polariton damping is graphically illustrated by Fig. 2.

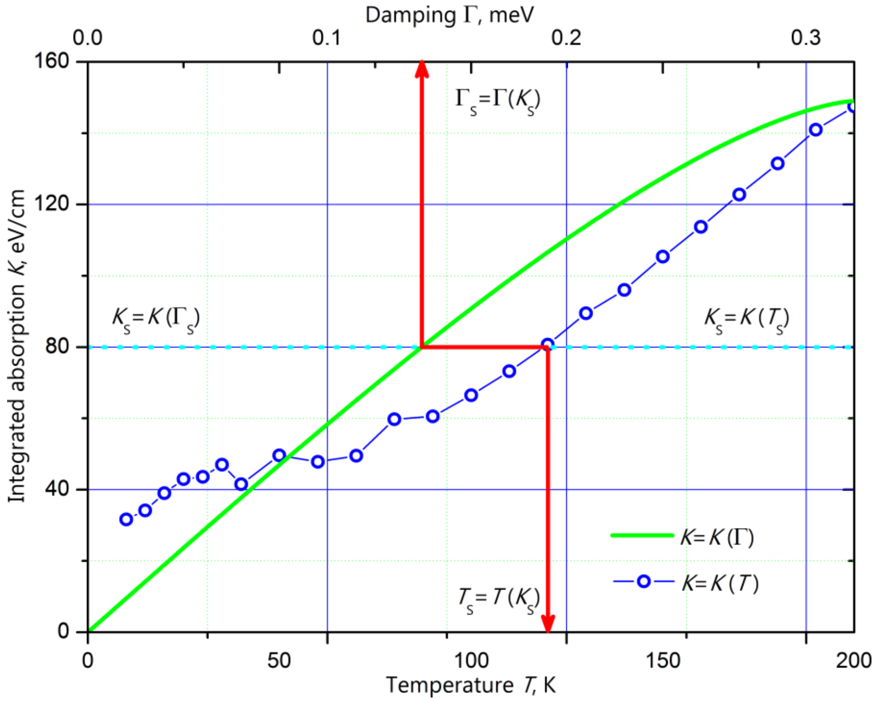

Figure 2. The graphical illustration of the extraction procedure of exciton-polariton damping excluding the contribution of inhomogenous broadening into the observed linewidth. The experimental dependence of integrated absorption $K(T)$ is shown by blue dots with a line connecting experimental points. The theoretical dependence of integrated absorption $K(\Gamma)$ is shown by a green line. The horizontal cyan dash line designates a sample value of integrated absorption K_S=80 eV/cm correlating damping $\Gamma_S(K_S)$ to temperature $T_S(K_S)$. The paof correlated $\Gamma_S(K_S)$ and $T_S(K_S)$ is connected by a red line.

The temperature dependence of experimentally observed FWHM of exciton ground state absorption line $H(T)$ and the extracted dissipative damping $\Gamma(T)$ are plotted at Fig. 3 on a logarithmic scale.

Analyzing the extracted temperature dependant damping $\Gamma=\Gamma(T)$ it is possible to determine material parameters regarding the main temperature dependent and non-temperature dependent dissipative mechanisms of exciton-polariton interaction with neutral and ionized impurities, acoustic and optical phonons Markosov (2009).

$$\Gamma(T) = \Gamma_0+\alpha\cdot T+\beta/[\exp(\hbar\Omega_{LO}/k_B T)-1] \quad (2).$$

where Γ_0 is the temperature-independent damping, α and β are material constants characterizing the scattering of exciton-polaritons by acoustic and optical phonons respectively

The α and β constants derived from the fits of the extracted damping curve (Fig. 3) matching the model (2) are listed in Table 1.

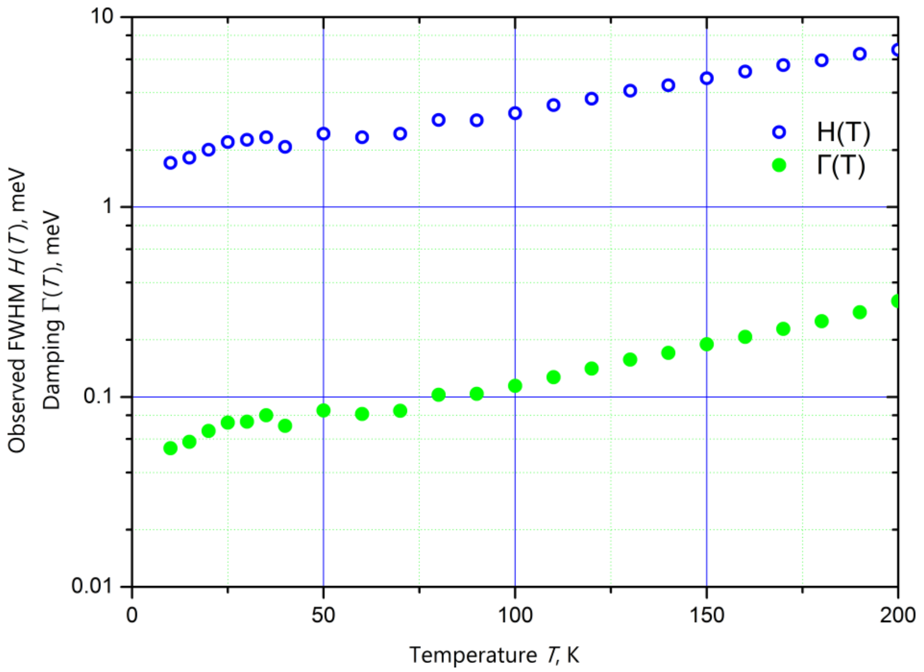

Figure 3. The temperature dependence of experimentally observed FWHM of exciton ground state absorption line $H(T)$ designated by blue dots and the extracted dissipative damping $\Gamma(T)$ designated by green dots, both plotted on a logarithmic scale.

Table 1. Experimentally obtained material parameters of investigated InP samples

Maximum integrated absorption K_{max}	149 eV/cm
Critical temperature T_c	200 K
Critical damping Γ_c	0.324 meV
Longitudinal-transverse exciton splitting $\hbar\omega_{LT}$	0.175 meV.
Temperature-independent damping Γ_0	0.0605 meV
Acoustic phonon scattering constant α	140 neV/K
Optical phonon scattering constant β	1.62 meV

Conclusions

At low temperatures below the critical one, the experimentally observed absorption linewidth of exciton ground state is substantially larger than the true dissipative damping. In this case, the linewidth is defined by the inhomogeneous broadening caused by imperfections and impurities in experimental samples.

The inhomogeneous broadening does not contribute into integrated absorption of exciton resonance and can be excluded when extracting the temperature dependence of exciton-polariton damping by the method integrated absorption. The extracted temperature dependence of the true damping was analyzed with regards to the main dissipative mechanisms.

The determined material parameters of exciton-polariton scattering by acoustic and optical phonons can be taken into account when developing prospective exciton-polariton devices.

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