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Electronic Supplementary Information: Tuning Exciton Diffusion, Mobility and Emission Line Width in CdSe Nanoplatelets via Lateral Size

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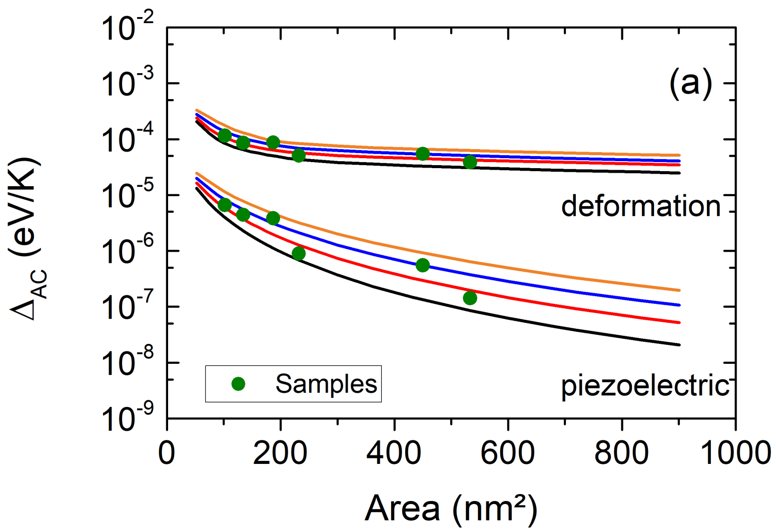
S1 Exciton-phonon coupling

A Coupling to acoustic phonons

In non-centrosymmetric crystals such as II-VI polar semiconductors, the coupling to acoustic phonons with linear dispersion $\omega_{\lambda,q} = c_\lambda q$ and sound velocity c_λ arises from both the deformation potential(DA) and LA as well as TA piezoelectric (PA) coupling mech-



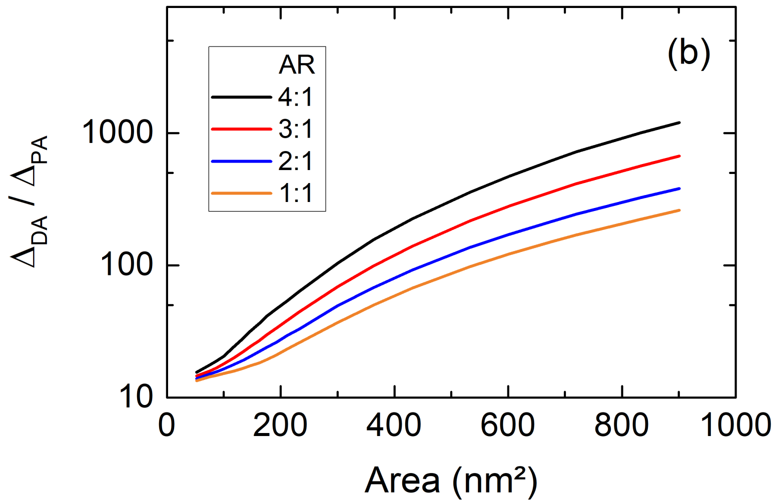


Figure S1: Theory: a) Coupling to deformation $\tilde{\Delta}_{DA}$ and piezoelectric $\tilde{\Delta}_{PA}$ interaction mechanisms versus lateral platelet area. b) Ratio of deformation potential to piezoelectic coupling $\tilde{\Delta}_{DA}/\tilde{\Delta}_{PA}$.

anisms.[1–4] The coupling matrix element therefore has contributions from both coupling mechanisms. So in order to emphasize the contribution of both coupling mechanisms to acoustic phonon interaction reported in Figure 2a of the main text, we display in Figure S1, the coupling to the deformation potential (top row) and piezoelectric (bottom row) interactions involving both LA and TA phonon, versus lateral platelet area and for different aspect ratios (AR), as obtained from theory (see Methods section). The two mechanisms have been obtained from the coupling matrix element in Eqs. (11) and (12) outlined in the method section and with the parameters listed in Table 1. We can clearly see in Fig. S1 (b) that the acoustic phonon scattering mechanisms are dominated by the deformation potential interaction rather than piezoelectric coupling (, so that for the scattering rates $\Gamma_{AC} \simeq \Gamma_{DA}$ is valid). In fact, it is found that the contribution of deformation potential coupling is more than one order of magnitude larger than the piezoelectric coupling for small lateral platelets sizes. It increases dramatically to about 3 orders of magnitude difference for larger platelets. Hence piezoelectric coupling in NPLs is a minor contribution.

B Coupling to optical phonons

The coupling to optical phonons may be mediated by unpolar (optical) zero order deformation-potential coupling (NP,Def) and Fröhlich interaction (LO,Fr).[1,4,5] In Fig. 2b of the main text, we have presented the coupling to optical phonon using both mechanisms, as calculated by our theory presented in the methods section. By comparing the results for unpolar (optical) zero order deformation-potential and Fröhlich coupling mechanism, we find that the polar optical phonon contribution via Fröhlich interaction is clearly dominant ($\Gamma_{Opt} \simeq \Gamma_{LO,Fr}$). The optical zero-order deformation potential contribution is weak, about 2 order of magnitude smaller than the Fröhlich interaction, see Figure S2. Hence it is nearly negligible. Notably, the simultaneous coupling via the two mechanisms in the optical and acoustic phonon interaction gives rise to interference between them when they are in phase. In our calculation, the coupling mechanisms involved in the optical and acoustic phonon interaction are often assumed to be out of phase; i.e., one is real and the other imaginary (see methods section). This implies that the two coupling mechanisms (piezoelectric and deformation coupling for the acoustic mode and the non polar deformation and polar Fröhlich coupling for the optical mode) do not interfere, and can therefore be treated as separate scattering mechanisms.

To illustrate the effect of the unpolar optical deformation potential and the piezoelectric potential on the total mobility presented in Fig 3 and 4 of the main text, we calculate in table S1 the total mobility $\frac{1}{\mu_{tot}} = \frac{1}{\mu_{LO,Fr}} + \frac{1}{\mu_{NP,Def}} + \frac{1}{\mu_{DA}} + \frac{1}{\mu_{PA}}$ using Matthiessens rule. In number of appearence the LO-phonon Fröhlich and optical deformation potential interaction as well as the acoustic deformation potential and pizoelectric interaction are

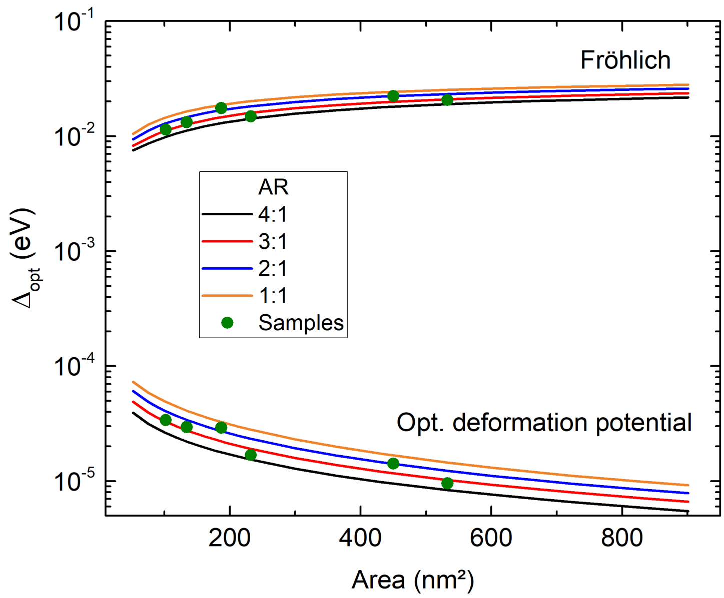


Figure S2: Coupling to unpolar (optical) zero order deformation-potential coupling $\tilde{\Delta}_{NP,Def}$ and Fröhlich interaction $\tilde{\Delta}_{LO,Fr}$ versus lateral platelet area

included in the expression. We compare the result with the mobility without unpolar optical phonon scattering (NP,Def) and/or piezo-accoustic scattering (PA) for two exemplary platelet sizes (a small and a large one) of 134 nm^2 and 533 nm^2 at two different temperatures (4 and 300 K). By comparing μ_{tot} with $\mu_{LO,Fr+DA+PA}$ in Table S1, we find that at both T=4K and 300 K the contribution of the unpolar optical phonon scattering is negligible due to the reciprocal summation to the total mobility according to Matthiesens rule. This confirms that optical phonon scattering is dominated by Fröhlich interaction (Fr).

In order to test the contributions of the piezoelectric (acoustic) interaction and optical deformation potential, we compare μ_{tot} with $\mu_{LO,Fr+DA+NP,Def}$ and $\mu_{LO,Fr+DA+PA}$.

Table S1: The mobility with and without the unpolar optical phonon coupling contribution and/or the piezoelectric coupling contribution for for two exemplary platelet sizes 134 nm^2 and 533 nm^2 at 4 and 300 K.

	134 nm^2		533 nm^2	
Mobility (cm^2/Vs)	4 K	300 K	4 K	300 K
$\mu_{LO,Fr+DA}$	$5.37\cdot10^3$	54.4	$11.9\cdot10^3$	76.0
$\mu_{LO,Fr+DA+NP,Def}$	$5.37\cdot10^3$	54.4	$11.9\cdot10^3$	76.0
$\mu_{LO,Fr+DA+PA}$	$5.11\cdot10^3$	52.4	$11.9\cdot10^3$	75.9
μ_{tot}	$5.11\cdot10^3$	52.3	$11.9\cdot10^3$	75.9

We can clearly see based on Table S1 that the inclusion of the piezoelectric interaction (PA) results a only slight reduction of the total mobility. Here, the deformation potential contribution (DA) to acoustic phonon scattering is significantly larger than the PA contribution.

On the other hand optical phonon scattering is dominated by Fröhlich interaction and the non-polar optical deformation potential coupling (NP.Def) results, in-line with Figure S2, in no (signifficant) alteration of the total mobility.

C Effect of σ on the optical phonon interaction

In the Fröhlich interaction, we use a σ value from Ref. 6 for CdSe nanoplatelets, which is also near to published values for TMDC materials,[5] see Table 1 of the Methods section. Since σ is not apriori well known and to illustrate the effect of an uncertainty in this parameter, we performed in Fig S3 a σ variation calculation, in order to investigate its effect on the optical phonon coupling and subsequently the impact on the uncertainty of the LO-coupling. We use also σ (apart from other parameter uncertainties taken into account) for estimating the contribution to the theoretical error bars in Fig. 2b.

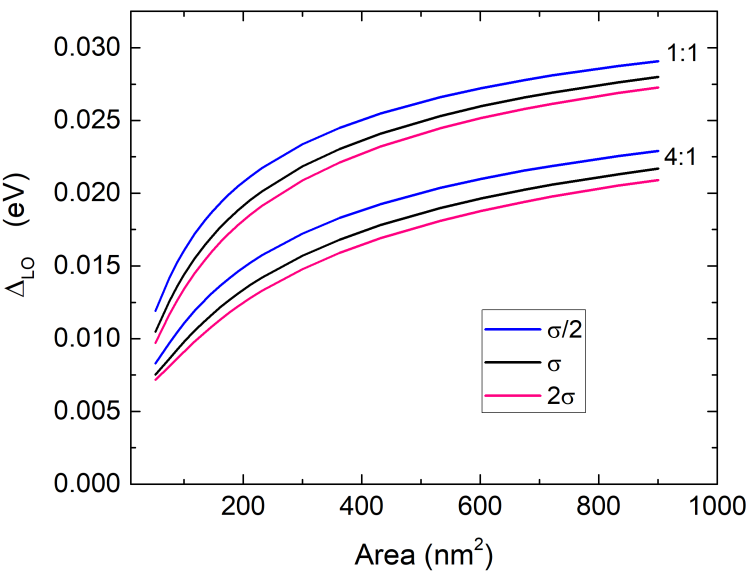


Figure S3: (a-d) The optical phonon coupling for a variation of σ (as used in Table 1, Methods section) versus lateral platelet area for 4:1 and 1:1 lateral aspect ratio.

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