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On binding energy of trions in bulk materials

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ABSTRACT

We study the negatively T^- and positively T^+ charged trions in bulk materials in the effective mass approximation within the framework of a potential model. The binding energies of trions in various semiconductors are calculated by employing Faddeev equation in configuration space. Results of calculations of the binding energies for T^- are consistent with previous computational studies and are in reasonable agreement with experimental measurements, while the T^+ is unbound for all considered cases. The mechanism of formation of the binding energy of trions is analyzed by comparing contributions of a mass-polarization term related to kinetic energy operators and a term related to the Coulomb repulsion of identical particles.

Excitonic effects in semiconductors are determined by the exciton binding energy and electron–hole interaction and play a critical role in optoelectronic devices [1]. Charged exciton complexes such as negative (T^-) and positive (T^+) trions are formed when a single exciton is correlated with an additional electron in a conduction band or hole in a valence band, respectively, has been proposed by Lampert [2]. In the meantime T^- and T^+ trions have been the subject of intense studies in the last two decades, both experimentally and theoretically. Their observation in bulk semiconductors has been hampered due to their rather small binding energies and became a challenging task. Trions were first observed in quantum wells [3] in 1993 and shortly thereafter in GaAs–AlGaAs quantum wells [4–6]. Trions were predicted and found in the photoluminescence and absorption spectra of various optically excited semiconductors, especially in quantum dots [7,8], quantum wells [1,9,10] and carbon nanotubes [11,12]. Mott–Wannier trions in two- and three-dimensional (2D and 3D) semiconductors can be described by the solutions of the three-body Schrödinger equation after modelling the crystal by effective electron and hole masses and a dielectric constant. There are stochastic variational calculations, and studies by using density functional theory, variational quantum Monte Carlo method, and the diffusion Monte Carlo approach [13–15]. Calculations have shown that the binding energy of a trion is strongly enhanced in two-dimensional structures due to the trion's larger spatial extent. Trions have been observed in 2D transition-metal dichalcogenide (TMDC) semiconductors [16–25]. Until now several approaches have been proposed for evaluations of the binding energies of a trion in two-dimensional transition metal dichalcogenides. Initial work on trion binding energies in TMDCs employed variational wave functions [26], and more recently using the time-dependent density-matrix functional theory, the fractional dimensional space approach, the stochastic variational method with explicitly correlated Gaussian basis, method of hyperspherical harmonics, and quantum Monte Carlo methods, such as the diffusion Monte Carlo and the path integral Monte Carlo [26–37]. Let us note that trions are studied in anisotropic two-dimensional materials such as phosphorene and arsenene [38], and are predicted to have remarkably high binding energies. Though much progress has been made, intrinsic excitonic states of 2D and 3D trions are still highly debated in theory, particularly related to the binding energies for negatively and positively charged trions which thirsts for experimental determination. In this letter we address this issue.

Because trions are intrinsically three-particle objects, common calculation methods are not always adequate to describe their behavior and a more rigorous level of theory must be employed. In the present work we study the T^- and T^+ trions within the Faddeev formalism [39], – the most rigorous approach for investigating a three-body system. In the case of a trion one deals with a three-body system AAB with two identical particles. We perform ground-state calculations for a positively and negatively charged trion in the effective mass approximation within the framework of a nonrelativistic potential model using the method of Faddeev equations in configuration space [39]. This approach gives new

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insights to the problem, because the Faddeev equations are the most general equations for description of a non-relativistic three-particle system within the potential approach and use as inputs only masses of particles and pairwise inter-particle interaction. There are no any fitting parameters in our approach. In the case of trions in bulk the inter-particle interaction is described by the Coulomb potential and the electron and hole masses can be obtained by different *ab initio* methods: many-body G_0W_0 and GW, density functional theory and the local density approximation. Therefore, one can understand what kind other quantum effects, which are not included in the potential model, should be considered for an adequate description of trions. In our approach, the trion is a three-particle system *eeh* (*hhe*) consisting of electrons (e) and heavy holes (h), with each pair interacting by the Coulomb force. To understand the origin of the difference of binding energies for the mirror systems of charged trions, we solve the Faddeev equations for cases when all three particles are interacting via the Coulomb potential and when the interaction between two identical particles is omitted or is screened and use these solutions to analyze contributions of two terms which course the difference of binding energies. The first term is related to the Coulomb repulsion between two identical particles and the second one is the mass-polarization term (MPT) [40] related to the kinetic energy operators. The latter term can be most clearly introduced by using the Schrödinger equation in the system of reference relative to the non-identical particle.

The Faddeev equations in configuration space can be written in the form of a system of second order differential equations [39], which can be reduced to a simpler form for the case of two identical particles. In this case the total wave function of the system is decomposed into the sum of the Faddeev components U and W corresponding to the $(AA)B$ and $(AB)B$ types of rearrangements: $\Psi = U + W \pm PW$, where P is the permutation operator for two identical particles. In the latter expression the sign "+" corresponds to two identical bosons, while the sign "−" corresponds to two identical fermions, respectively. After introducing the set of the Jacobi coordinates for the three particles, separating the motion of the center-of-mass one can write the set of Faddeev equations for the relative motion of three particles when two of them are identical fermions in the following form [43,44]:

$$\begin{aligned}(H_0 + V_{AA} - E)U &= -V_{AA}(W - PW),\\ (H_0 + V_{AB} - E)W &= -V_{AB}(U - PW).\end{aligned} \tag{1}$$

In Eq. (1) the Hamiltonian H_0 is the operator of kinetic energy written in terms of corresponding Jacobi coordinates, while V_{AA} and V_{AB} are the potentials of the pairwise interactions between the particles. The pairwise interactions are described by the Coulomb potential with the dielectric constant related to the considered material.

Let us consider the states of T^- and T^+ trions with the total angular momentum $L = 0$, the momentum of pair $l = 0$, and momentum $\lambda = 0$ of the third particle with respect to the center-of-mass of the pair. Within this condition the pair of electrons (holes) is in a singlet spin state. The corresponding spin function is asymmetric with respect to the permutation operator P, which provides automatically the asymmetry of the trion wave function Ψ: $P\Psi = P(U + W - PW) = -U + PW - W = -\Psi$.

To analyze the origin of the difference of binding energies for the T^- and T^+ trions let us follow Ref. [40] and write the Schrödinger equation for the trion in the system of reference relative to the non-identical particle:

$$\begin{aligned}(-\frac{\hbar^2}{2\mu}\nabla^2_{r_{A_1}} - \frac{\hbar^2}{2\mu}\nabla^2_{r_{A_2}} - \frac{\hbar^2}{m_B}\nabla_{r_{A_1}}\nabla_{r_{A_2}} + V_{AB}(r_{A_1}) + V_{AB}(r_{A_1})\\ - V_{AA}(r_{A_1} - r_{A_2}) - E_3)\Psi(r_{A_1}, r_{A_2}, r_{A_1} - r_{A_2}) = 0,\end{aligned} \tag{2}$$

which is written in a self-explanatory notation. In Eq. (2) μ is the reduced mass of the electron and hole and $T_{mp} = -\frac{\hbar^2}{m_B}\nabla_{r_{A_1}}\nabla_{r_{A_2}}$ is the mass-polarization term and E_3 is the ground state energy of the three particles. In the case $m_B < m_A$ the contribution of the MPT can be of the same order as the contribution of the other two differential operators in Eq. (2) due to the comparable mass factors of these operators, which can be expressed as $1/m_B$. In the case $m_B > m_A$ the contribution of this term has the factor $1/m_B$, while the mass factors of other differential operators are of the order of $1/m_A$. When $m_B >> m_A$ the contribution of the MPT can be ignored. If in Eq. (2) the MPT and the interaction $V_{AA} \equiv V_{AA}(r_{A_1}, r_{A_2}, r_{A_1} - r_{A_2})$ between two identical particles are neglected one obtains:

$$\begin{aligned}(-\frac{\hbar^2}{2\mu}\nabla^2_{r_{A_1}} - \frac{\hbar^2}{2\mu}\nabla^2_{r_{A_2}} + V_{AB}(r_{A_1}) + V_{AB}(r_{A_1})\\ - E_3(T_{mp} = 0, V_{AA} = 0))\Psi(r_{A_1}, r_{A_2}) = 0,\end{aligned} \tag{3}$$

where $E_3(T_{mp} = 0, V_{AA} = 0)$ is the ground state energy of the three particle system for the aforementioned condition. In Eq. (3) the total wave function can be factorized as $\Psi(r_{A_1}, r_{A_2}) = \Phi(r_{A_1})\Phi(r_{A_2})$, where $\Phi(r_{A_1})$ is a solution of two-body Schrödinger equation for the AB subsystem, and that leads to the trivial solution: $E_3(T_{mp} = 0, V_{AA} = 0) = 2E_2$, where E_2 is the ground state energy of two-body subsystem AB. Within our consideration the bound AB pair is the exciton.

To evaluate the effect of the MPT, one can neglect the interaction between the identical particles, $V_{AA} = 0$, in Eq. (2). The evaluation can be written as $\Delta = E_3(T_{mp} = 0, V_{AA} = 0) - E_3(V_{AA} = 0)$, where $E_3(V_{AA} = 0)$ is the ground state energy of three particle system AAB when interaction between the identical particles is neglected.

Taking into account Eq. (3), the last expression can be rewritten as

$$\Delta = 2E_2 - E_3(V_{AA} = 0) = B_3(V_{AA} = 0) - 2B_2 \geq 0, \tag{4}$$

where B_2 and $B_3(V_{AA} = 0)$ are the binding energies of the exciton and three-body AAB system when the interaction between two identical particle is omitted, respectively. In the simplest case when $m_B >> m_A$ the contribution of the MPT can be neglected and one has: $B_3(V_{AA} = 0, < T_{mp} >= 0) = 2B_2$. In consequence, Eq. (4) is valid for any mass ratio m_B/m_A and can be used to evaluate the effect of the mass polarization term. The relation (4) is known in nuclear physics as the mass polarization effect [40–42].

Let us introduce the interaction between two identical particles as $\alpha < V_{AA} >$, where the parameter α controls the strength of this interaction. Substituting this potential in Eq. (2) and averaging it gives the following expression:

$$\begin{aligned}E_3 = &- < T_1 > - < T_2 > + < V_{AB} > + < V_{AB} > - < T_{mp} >\\ &+ \alpha < V_{AA} > .\end{aligned} \tag{5}$$

In Eq. (5) the matrix elements $< T_1 > = < T_2 >$ and $< V_{AB} > = < V_{AB} >$ due to the symmetry of the system. Thus, from the one hand by solving of the Faddeev equations (1) one can find binding energies B_3 for the T^- and T^+ trions and test the sensitivity of their binding energy to the strength of $\alpha < V_{AA} >$ by varying the parameter α. On the other hand, by solving (1) under the condition $V_{AA} = 0$, one can find the binding energies $B_3(V_{AA} = 0)$ for trions when the interaction between two identical particles is omitted. Such an approach allows one to analyze the origin of the binding energy difference for T^- and T^+ trions. Obviously, $< T_{mp} >$ is related to Δ as well as $< T_{mp} > \approx \Delta$ when the contribution of MPT to $E_3(V_{AA} = 0)$ is small.

Table 1
The charged and neutral exciton binding energy deference $B_T = B_3 - B_2$ (the binding energy of trion with respect to the exciton binding energy) for different materials. B_2 and B_3 are the binding energy for the trion and exciton, respectively. The relative contribution of the mass polarization term δ is calculated as $\delta = (B_3(V_{AA}=0) - 2B_2)/B_3(V_{AA}=0)$. Here $\hbar^2/m_0 = 7.6195$ eV Å^2, ϵ is the dielectric constant. All energies are given in meV and the masses are given in units of a free electron mass m_0.

Material: m_e, m_h, ϵ	m_h/m_e	m_e/m_h	Trion	B_2	B_3	$B_3(V_{AA}=0)$	B_T	δ
InN: 0.11, 1.63, 7.5 [45]	14.8		T^-	24.88	28.5	49.8	3.6	~0%
		0.07	T^+	24.88	–	65.8	–	24%
GaAs: 0.067, 0.51, 12.9	7.6		T^-	4.83	5.33	9.66	0.5	~0%
		0.13	T^+	4.83	–	11.8	–	18%
ZnSe: 0.16, 0.75, 8.6	4.7		T^-	24.2	26.3	48.4	2.1	~0%
		0.2	T^+	24.2	–	55.7	–	13%
GaN: 0.2, 0.82, 8.9	4.1		T^-	27.57	29.6	55.1	2.1	~0%
		0.24	T^+	27.57	–	62.4	–	12%
CdTe: 0.096, 0.35, 10.16	3.6		T^-	9.91	10.6	19.8	0.6	~0%
		0.27	T^+	9.91	–	22.2	–	11%
MoS_2: 0.45, 0.45, 12.6 [50]	1.0		T^-	19.3	–	39.3	–	1.7%
		1.0	T^+	19.3	–	39.3	–	1.7%
0.45, 0.45, 10.7 [51,52]	1.0		T^-	26.7	26.8	54.5	~0.1	2.0%
		1.0	T^+	26.7	26.8	54.5	~0.1	2.0%

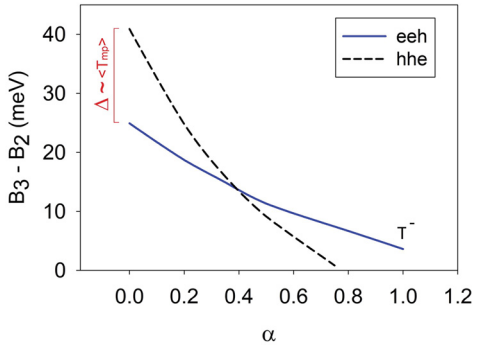

Fig. 1. The $B_3 - B_2$ for *eeh* (solid curve) and *hhe* (dashed curve) in InN as a function of the scaling factor α of the Coulomb repulsion between identical particles. The crossing point corresponds to $\alpha = 0.41$. The relative value of the mass polarization term $< T_{mp} >$ for *hhe* is shown by the vertical line segment.

Let us discuss results of calculations for excitons and $T^{\pm}$ trions binding energies in various bulk semiconductors. The binding energies B_3 and $B_3(V_{AA}=0)$ for the trions were calculated using the aforementioned Faddeev formalism. We calculate the binding energy B_2 of the exciton in the same semiconductor as well. In our calculations we use the known mass ratio m_B/m_A for various bulk materials and the corresponding dielectric constant ϵ. The numerical results of our calculations presented in Table 1 show that the negative trion is always bound, while the positively charged trion has no bound state for all set of parameters. To demonstrate that for the particular equal masses the negatively and positively charged trions have the same energies and can be bound or unbound depending on the value of the dielectric constant we perform calculations for trions binding energies in bulk MoS_2 which are presented in Table 1. The analyses of the results for $B_3(V_{AA}=0)$ and binding energy of the exciton B_2 shows that the relation $B_3(V_{AA}=0) > 2B_2$ is always satisfied for trions and relative contribution of the MPT indicated by $\delta = (B_3(V_{AA}=0) - 2B_2)/B_3(V_{AA}=0)$. For the positively charged trions, δ increases when the ratio m_B/m_A decreases. For the negatively charged trions, the MPT effect is small and $\delta \sim 0$.

Let us mention that the results for B_2 confirm the hydrogenic exciton energy $B_2 = Ry^* = 13.61\mu^*/\epsilon_r^2$ (eV) (Ry^* is the effective Rydberg constant with the reduced electron–hole effective mass μ^*) and are in good agreement with experimental data and theoretical calculations [10,15,45–49]. Fig. 1 presents the difference $B_3 - B_2$ for AAB systems as a function of α, which controls the strength of interaction between identical particles: $V_{AA} \to \alpha V_{AA}$. The difference between the curves at $\alpha = 0$ shows the contribution of MPT for the *hhe* system relative to the *eeh*. The slopes of the curves differ significantly. This indicates that the repulsion between identical particle is much stronger in the *hhe* system than in *eeh* because the two holes are localized much closer to each other due to their larger effective masses. Thus, our hypothetical model with parameter α, which controls the strength of interaction between identical particles for both trions and effectively represents screening caused predominantly by the valence electrons, leads to a weaker Coulomb interaction between the identical particles and hence an increased trion binding energy. However, this interaction is stronger for two holes because they are localized more closely to each other than electrons, due to the larger kinetic energy caused by the heavier effective mass of holes, and hence a reduced trion binding energy. Therefore, the effect of strong repulsion due to the Coulomb interaction takes place. This fact is illustrated in Fig. 2 by the contour plots of the Faddeev component U and W: the negative trion has more extended distribution within about 200 × 250 Å, than less extended distribution within 80 × 120 Å for the positive trion. The analyses of Figs. 1 and 2 allows to conclude that both systems are bound with the same binging energy when $\alpha = 0.41$, However, the *hhe* is more compact. One can make the transformation of T^- to T^+ by replacing of the masses of the electron and hole and vice versa ($m_e \to m_h$ and $m_h \to m_e$). The parameter $\xi \geq 0$ sets this transformation as follows: $m_A^{\xi} = (1+\xi)m_A, m_B^{\xi} = (1 - \xi m_A/m_B)m_B$ and keeps the sum of the masses, $m_A + m_B = const$. For example, for the effective masses of the electron and hole in InN, when $\xi = 0$ we have T^-, while for $\xi = 13.75$ we have T^+. The dependence of different characteristics for *eeh* and *hhe* systems as a function of the parameter ξ is presented in Fig. 3. During the transformation, the energy E_3 follows B_2, does not return to the initial value defined for $\xi = 0$ due to the strong increasing of the Coulomb contribution (see Fig. 3a)). The contribution of the Coulomb repulsion increases more quickly than the B_2 when the ratio m_B/m_A increases.

The replacement $e \to h$ in the *eeh* system, when the AA interaction is omitted, leads to the increase of the MPT contribution indicated by the increasing $B_3(V_{AA}=0)$ relatively $2B_2$ (Fig. 3b)). Note that, in case of GaAs, the relative increase varies from 0% to 18% or, in absolute values, from 9.66 meV to 11.8 meV.

The analyses of the results presented in Fig. 3 shows that the matrix element of the Coulomb repulsion $< V_{AA} >$ increases more quickly than the MPT, $< T_{mp} >$. In another words, small variations of the masses (making more compactness of the system) generate

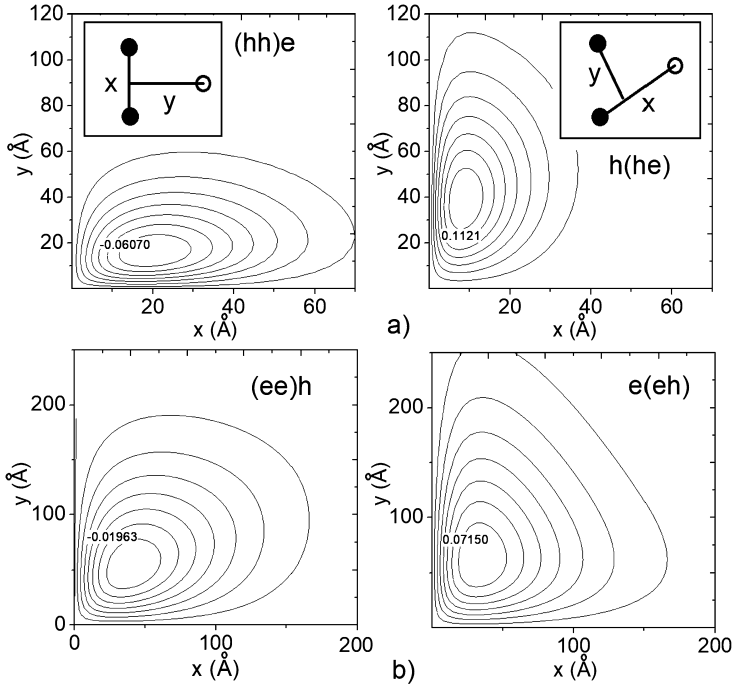

Fig. 2. The contour plots of the Faddeev components U (left) and W (right) for a) *hhe* and b) *eeh* systems when the scaling factor for the *ee* (or *hh*) Coulomb repulsion $\alpha = 0.41$ (see Fig. 1). The inserts show the corresponding Jacobi trees and Jacobi coordinates x and y are measured in Å.

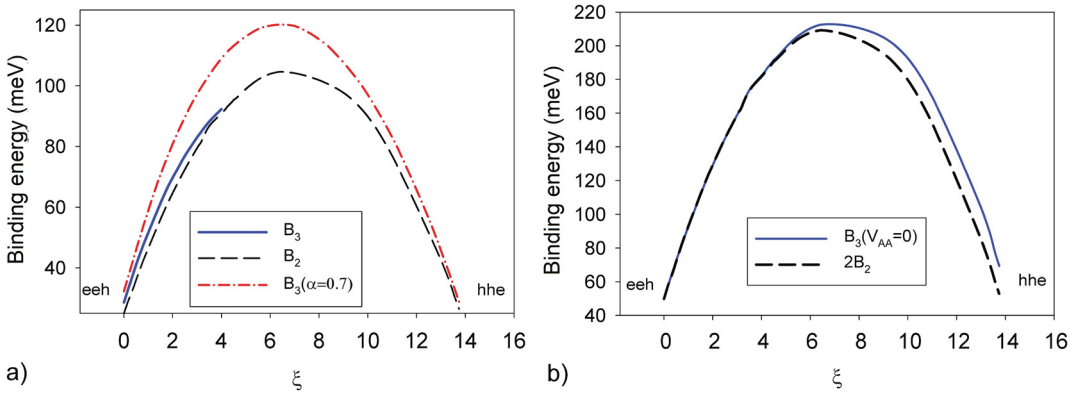

Fig. 3. The mass transformation from *eeh* to *hhe* in InN. a) The binding energies B_3 (solid curve), B_2 (dashed curve) and $B_3(\alpha = 0.7)$ (dot-dashed curve); and b) $2B_2$ (dashed curve), and $B_3(V_{AA} = 0)$ (solid curve), as a function of the mass transformation parameter ξ. The parameter ξ is related to the negative trion, when $\xi = 0$, and to the positive trion, when $\xi = 13.75$.

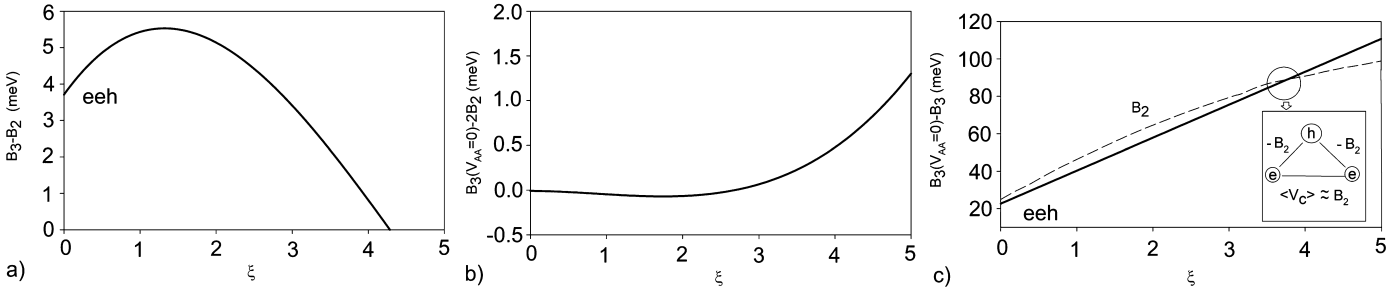

Fig. 4. The value of a) $B_3 - B_2$, b) $B_3(V_{AA} = 0) - 2B_2$, c) $B_3 - B_3(V_{AA} = 0)$ for *eeh* in InN as a function of the mass transformation parameter $\xi \leq 5$ (solid curve). The B_2 is shown by the dashed curve. The inset shows schematic distribution for energies in the system.

larger Coulomb repulsion between identical particles. The latter results in the unbound state of the transforming *eeh* system and the final *hhe* system (T^+).

One can reduce this Coulomb repulsion artificially using the parameter α. The results are presented in Fig. 3a). The energy of T^+ is evaluated as 0.7 meV above the *eh* threshold when $\alpha = 0.7$. In this case, E_3 is larger B_2 during the transformation and in the final point. It means also that the MPT can compensate for the increased Coulomb repulsion.

The Coulomb repulsion of AA particles can be evaluated as $\Delta B_c(AA) = B_3 - B_3(V_{AA} = 0)$. This is shown in Fig. 4, which presents the dependence of different characteristics for the *eeh*

system on the parameter $\xi = 0$. One can conclude that, firstly, the binding energy of *eeh* system $B_T = B_3 - B_2$, $B_T/B_2 << 1$ decreases during the mass transformation to the negative value (unbound state) when $\xi \sim 4$ (Fig. 4 a)). One can further conclude, secondly, the contribution of the MPT increases with the increment of ξ and hence when the mass ratio m_B/m_A increases. However, the MPT related additional energy is small relative to B_T and cannot compensate for the Coulomb repulsion. Thirdly, the contribution of the Coulomb repulsion for T^- near the point $\xi \sim 4$ can be evaluated as $< V_c > \sim B_2$ taking into account the small binding energy of T^-, as well as $B_T/B_2 << 1$. The latter means that the *eeh* system is clustered as $e + (eh)$ and the energy separation of the electron from the pair *eh* is small. According to Eq. (5) one can writes: $B_3 = 2B_2 - < V_c > \approx B_2$ and $< V_c > \sim B_2$.

We have shown that the matrix element $< V_c >$ of the Coulomb interaction for two identical particles in the *hhe* system is larger than one in the *eeh* system. When one disregards the V_{AA} interaction, the large MPT in the *hhe* makes the system more bounded compares with the *eeh* system, where the contribution of the MPT term is negligible. Using scale factor $\alpha \geq 0$ for the AA interaction we show that the relation $< V_c(hhe) > \geq < V_c(eeh) >$ is satisfied for increasing α up to $\alpha \approx 1$. The system *hhe* becomes unbound rapidly when α is increasing. According to Eq. (5) one can write: $B_3 = 2B_2 + < T_{mp} > -\alpha < V_c >$, and can see that $B_3 \approx B_2$ when the screening is defined by $\alpha \approx 0.7$. The binding energies of the $T^{\pm}$ trions are calculated for different bulk materials based on the Faddeev equation for the AAB system in configuration space. It was found that the binding energy of T^- is relatively small, $B_T/B_2 << 1$, while T^+ is unbound. The results of the calculations for B_T are consistent with previous computational studies and are in reasonable agreement with experimental measurements. We explain the origin of the difference of binding energies of T^- and T^+ by using the Schrödinger equation written in the system of reference relative to the non-identical particle. There are two terms of the equation which play an important role for the formation of the bound state of a trion when $m_e/m_h << 1$: the Coulomb repulsion between two identical particles and MPT. The MPT, T_{mp}, adds a part to the binding energy of the AAB system, while the Coulomb repulsion between AA identical particles decreases the energy. Comparing the bound and unbound states of $T^{\pm}$ in considered materials, we show that hole–hole Coulomb repulsion is stronger in T^+ than the electron–electron one in T^- due to more close localization of the two holes. The last condition is possible due to large contribution of the MPT. By introducing the scaling parameter $0 \leq \alpha \leq 1$ and calculation of the binding energy as a function of α, we show that $< V_{AA} >$ is larger for the hole–hole pair.

We illustrated the interplay of these two terms by the hypothetical mass transformation $eeh \rightarrow hhe$, which replaces of the masses of the electron and the hole in bulk InN. Using this transformation we demonstrate that the Coulomb repulsion decreases more quickly and the MPT contribution, $< T_{mp} >$, does not compensate for the binding energy decrease. It was demonstrated that T^+ can be bound by reducing the strength of the Coulomb repulsion with the controlled parameter $\alpha < 1$.

The properties of the *eeh* and *hhe* systems are similar for 3D and 2D models. The similarity is based on the existence of the MPT and the Coulomb repulsion in the Schrödinger equation for the both cases. As we show, the interplay of the terms results in the bound or unbound state of the systems.

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