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On the possibility of self-trapping transition of acoustic polarons in two dimensions*

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A 2D electron-longitudinal-acoustic-phonon interaction Hamiltonian is derived and used to calculate the ground-state energy of the acoustic polarons in two dimensions. The numerical results for the ground-state energy of the acoustic polarons in two and three dimensions are obtained. The 3D results agree with those obtained by using the Feynman path-integral approach. It is found that the critical coupling constant of the transition from the quasifree state to the self-trapped state in the 2D case is much smaller than in the 3D case for a given cutoff wave-vector. The theory has been used to judge the possibility of the self-trapping for several real materials. The results indicate that the self-trappings of the electrons in AlN and the holes in AlN and GaN are expected to be observed in 2D systems.

Keywords: acoustic polarons, self trapping, two-dimensional electron-phonon interaction
PACC: 7138, 7145, 6320K

1. Introduction

The self-trapping of an electron in a deformable lattice has been maintaining interest of many scientists in the past decades.[1−13] Various calculations of the ground-state energies for acoustic polarons as functions of the electron-acoustic-phonon coupling strength have led to a transition from the quasifree state to the self-trapped state.[2−9] This transition phenomenon is also called 'phase transition', though it is not a real phase transition in general.[8] As is well known, the self-trapping states of electrons are more advantageous in the sense of energy than the wide band Bloch states.[12] Therefore the investigations of the self trapping of electrons are expected to be helpful for exploring the characteristics of materials, in particular, for lower-dimensional structures.[14−16]

An electron interacts with the acoustic and optical modes of the lattice vibration in a polar crystal. However, the abrupt change of the polaron state from the quasi-free state to the self-trapping state is usually caused by the short-range acoustic interaction, i.e. the electron-longitudinal-acoustic-phonon (e-LA-p) coupling, but not by the long-range longitudinal-optical (LO)-phonon interaction.[3] Therefore the acoustic polarons and their self-trapping have been attracting much attention theoretically in recent years.[17−20]

Some authors have discussed the self-trapping transition of acoustic polarons in three-dimensional (3D), two-dimensional (2D) as well as one-dimensional (1D) systems pure theoretically in early years.[3−7,12] It has been pointed out that a 3D acoustic polaron is impossible to be trapped in most semiconductors and III-V compounds, even in alkali halides.[5] Otherwise, the electron–phonon (e-p) coupling effects would be substantially enhanced in confined structures, such as quasi-2D and 1D systems, so that the self trapping transition would be easier to realize.[4,6,7,12] Therefore a reasonable lower 'transition point' of the e-p coupling strength from the quasi free state to the trapped state should be obtained theoretically for low-dimensional systems. Farias *et al* studied 2D acoustic polarons by using the Feynman path-integral formalism.[6] However, they obtained a transition point α_c which is higher than that in 3D systems and independent of cut-off wave-vector q_0. It might be concluded from the calculated results that the self-trapping of the 2D acoustic polarons are more difficult than the 3D ones. These conclusions are contrary to those given by Shoji and Tokuda.[4] It should be pointed out that the previous authors generally used the 3D-like e-p coupling Hamiltonian to describe 2D systems, which maybe underestimate the effect of the acoustic phonons on the 2D electronic states. Moreover the direct observations of the electron self-trapping in lead halides and the investigations of the

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effects on the photographic property by trapped electrons in silver halide recently have brought the refreshing breath into this field.[21−25] Therefore the further theoretical studies on the possibility for the self-trapping of acoustic polarons in low-dimensional and bulk materials are being carried out.

In this paper, we study the 'phase transition' of the 2D acoustic polarons from the quasi free state to the trapped state. For comparison, the self-trapping transition in 3D systems is also discussed. A Hamiltonian describing the 2D e-LA-p interaction is derived. The ground-state energy of the 2D acoustic polarons is calculated by using a Huybrechts-like variational approach.[26] The numerical results of the ground-state energies and their first-order derivatives for the acoustic polarons in the 2D and 3D systems are given as functions of the bulk e-LA-p coupling constant, and the possibility of the self-trapping is discussed. Finally we apply the theory to the study of some real materials.

2. 2D e-LA-p interaction Hamiltonian

The e-LA-p system in the 3D case is generally described by the following Hamiltonian:

$$H = \frac{P^2}{2m} + \sum_k \hbar\omega_k a_k^\dagger a_k + \sum_k \left(V_k a_k \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} + V_k^* a_k^\dagger \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} \right). \quad (1)$$

Here the linear dispersion $\omega_k = ck$ is used for the frequencies of acoustic phonons with a finite cutoff wave-vector k_0, and c is the speed of sound. The last term on the right-hand side in expression (1) is the Fröhlich Hamiltonian of the e-LA-p interaction.[3−5] The e-p coupling function is given by

$$V_k^* = \mathrm{i}\left(\frac{4\pi\alpha}{\Omega}\right)^{1/2} \frac{\hbar^2}{m} k^{1/2}, \quad (2)$$

where Ω is the volume of the crystal, and the dimensionless bulk e-LA-p coupling constant, α, is given by

$$\alpha = \frac{D^2 m^2}{8\pi\rho\hbar^3 c} \quad (3)$$

with D and ρ being the deformation potential the mass density of the crystal respectively.

Now we consider a 2D system, where an electron is confined in a 2D (x-y) plane of $z = 0$ and couples with LA-phonons. To analyse the possibility of the self-trapping of acoustic polarons in the 2D case, we have to seek for a reasonable e-LA-p interaction Hamiltonian for the system. A similar question is solved for 2D optical polarons in Refs.[27, 28]. For this purpose we divide a wave-vector of the phonon mode into the xy- and z-components, i.e. $\boldsymbol{k} \equiv (\boldsymbol{q}, k_z)$.[28,29] The e-p interaction Hamiltonian in expression (1) is then rewritten as

$$H_I = \sum_{q,k_z} (V_{q,k_z} a_{q,k_z} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{\rho}} + V_{q,k_z}^* a_{q,k_z}^\dagger \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{\rho}}), \quad (4)$$

where $\boldsymbol{\rho}$ is the 2D position-vector of the electron in the x-y plane.

To obtain the 2D e-LA-p interaction Hamiltonian, we sum $|V_{q,k_z}|^2$ over the z-component k_z of the phonon wave-vector with the cut-off wave-vector q_0. The 2D coupling coefficient then can be obtained as

$$|S_q|^2 = \sum_{k_z} |V_{q,k_z}|^2 = \sum_{k_z} \left(\frac{4\pi\alpha}{\Omega}\right) \frac{\hbar^4}{m^2} \sqrt{q^2 + k_z^2} = \frac{2\alpha}{A} \frac{\hbar^4}{m^2} \left[\frac{q_0^2}{2} \sqrt{1 + \frac{q^2}{q_0^2}} + \frac{q^2}{2} \ln \left(\frac{q_0}{q} + \sqrt{1 + \frac{q_0^2}{q^2}} \right) \right]. \quad (5)$$

In the above calculation the sum $\sum_{k_z}$ over k_z has been replaced by the integral $[L/(2\pi)]\int \mathrm{d}k_z$ with the upper-limit q_0, the boundary of the first Brillouin zone. A is the area of the 2D plane, and $\Omega = AL$.

The 2D e-LA-p system Hamiltonian is then written as

$$H = \frac{P^2}{2m} + \sum_q \hbar\omega_q a_q^\dagger a_q + \sum_q S_q^* a_q^\dagger \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{\rho}} + \mathrm{h.c.} \quad (6)$$

with

$$S_q^* = \mathrm{i}\left(\frac{2\alpha}{A}\right)^{1/2} \frac{\hbar^2}{m} \left[\frac{q_0^2}{2} \sqrt{1 + \frac{q^2}{q_0^2}} + \frac{q^2}{2} \ln \left(\frac{q_0}{q} + \sqrt{1 + \frac{q_0^2}{q^2}} \right) \right]^{1/2}. \quad (6a)$$

3. Variational treatment

Now we start with Hamiltonian (6) to calculate the ground-state energy of the 2D acoustic polaron, by using a Huybrechts-like variational approach.[26]

Firstly we carry out a unitary transformation

$$U_1 = \exp\left(-\mathrm{i}a\sum_q \boldsymbol{q}\cdot\boldsymbol{\rho}a_q^\dagger a_q\right), \tag{7}$$

where a is a variational parameter and will tend to 0 in a strong coupling limit, and 1 in a weak coupling limit. By doing so, the Hamiltonian turns into

$$H_1 = \frac{1}{2m}\left(\boldsymbol{p} - a\sum_q \hbar\boldsymbol{q}a_q^\dagger a_q\right)^2 + \sum_q \hbar\omega_q a_q^\dagger a_q + \sum_q \left[S_q^* a_q^\dagger \exp\left(-\mathrm{i}\left(1-a\right)\boldsymbol{q}\cdot\boldsymbol{\rho}\right) + \mathrm{h.c.}\right]. \tag{8}$$

Then we introduce linear combination operators of position and momentum of the electron respectively by using the following relations:

$$p_j = \left(\frac{m\hbar\lambda}{2}\right)^{1/2}\left(b_j^\dagger + b_j\right) \tag{9a}$$

and

$$\rho_j = \mathrm{i}\left(\frac{\hbar}{2m\lambda}\right)^{1/2}\left(b_j - b_j^\dagger\right) \tag{9b}$$

where $b_j^\dagger$ and b_j are respectively the creation and annihilation operators, $j = x, y$; λ is another variational parameter.

By inserting expressions (9a) and (9b) into expression (8) and performing the second unitary transformation

$$U_2 = \exp\sum_q \left(f_q a_q^\dagger - f_q^* a_q\right), \tag{10}$$

the Hamiltonian finally becomes the following form:

$$\begin{aligned} H_2 = {} & \frac{\hbar\lambda}{2}\left(\sum_j b_j^\dagger b_j + 1\right) + \sum_q \left(\hbar\omega_q + a^2\frac{\hbar^2q^2}{2m}\right)\left(a_q^\dagger a_q + f_q^* a_q + f_q a_q^\dagger + |f_q|^2\right) \\ & + \sum_q \left\{\left(S_q^* a_q^\dagger + S_q^* f_q^*\right)\exp\left[-\frac{\hbar\left(1-a\right)^2 q^2}{4m\lambda}\right]\exp\left[-\left(1-a\right)\left(\frac{\hbar}{2m\lambda}\right)^{1/2}\sum_j q_j b_j^\dagger\right]\right. \\ & \left.\times\exp\left[\left(1-a\right)\left(\frac{\hbar}{2m\lambda}\right)^{1/2}\sum_j q_j b_j\right] + \mathrm{h.c.}\right\} + \frac{a^2}{2m}\left(\sum_q \hbar q\left|f_q\right|^2\right)^2 - 2a\sum_{qj}\frac{\hbar q_j P_j}{2m}\left|f_q\right|^2. \end{aligned} \tag{11}$$

Here we have omitted multi-phonon processes, which contribute less to the polaronic energy.

The displacement amplitude in the second unitary transformation is determined as

$$f_q = -\frac{S_q^*\exp\left[-\hbar\left(1-a\right)^2 q^2/4m\lambda\right]}{\hbar\omega_q + a^2\hbar^2q^2/2m}, \tag{12}$$

by diagonalizing the vital important part of H_2.

The ground-state energy can be calculated by averaging Hamiltonian (11) over the zero-phonon state $|0\rangle$ of the acoustic polaron. Therefore we have

$$b_j\left|0\right\rangle = a_q\left|0\right\rangle = 0.$$

Through some standard treatments, the variational energy of the polaronic ground-state can be obtained as follows:

$$E_0 = \left\langle 0\right| H_2 \left|0\right\rangle = \frac{1}{2}\hbar\lambda\left(1-a\right)^2 + \sum_q\left(\hbar\omega_q + a^2\frac{\hbar^2q^2}{2m}\right)\left|f_q\right|^2 + \sum_q\left[S_q^* f_q^*\exp\left(-\frac{\hbar\left(1-a\right)^2q^2}{4m\lambda}\right) + \mathrm{h.c.}\right]. \tag{13}$$

Using expression(12) with $\omega_q = cq$ and replacing the summation $\sum_q$ by the integral $[A/(4\pi^2)]\int q\mathrm{d}q$ with the upper-limit q_0, the variational energy for the ground-state of acoustic polarons in the 2D case is finally written as

$$E_0 = \frac{1}{2}\hbar\lambda(1-a)^2 - \frac{\alpha}{\pi}\int_0^{q_0}\frac{\hbar cQ}{1+a^2q/2}\exp\left[-\frac{\hbar}{2m\lambda}(1-a)^2q^2\right]\mathrm{d}q, \tag{14}$$

where

$$Q = \frac{q_0^2}{2}\sqrt{1+\frac{q^2}{q_0^2}} + \frac{q^2}{2}\ln\left(\frac{q_0}{q}+\sqrt{1+\frac{q_0^2}{q^2}}\right). \tag{14a}$$

In expression (14) the variational parameters a and λ will be determined by numerically minimizing the energy in the following section.

For comparison, we have also calculated the self-trapping transition in 3D systems. The corresponding acoustic polaronic ground-state energy can be similarly obtained as

$$E_0 = \frac{3}{4}\hbar\lambda(1-a)^2 - \frac{2\alpha}{\pi}\int_0^{q_0}\frac{\hbar cq^2}{1+a^2q/2}\times\exp\left[-\frac{\hbar}{2m\lambda}(1-a)^2q^2\right]\mathrm{d}q, \tag{15}$$

by using Hamiltonian (1).

4. Numerical results and discussions

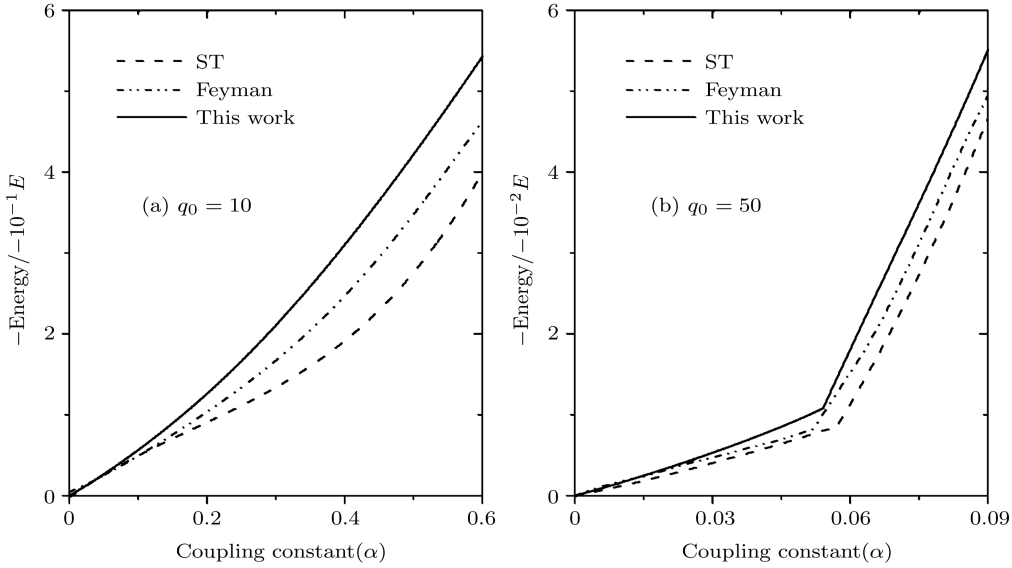


Fig.1. Ground-state energies of the acoustic polarons in the 3D case as functions of the e-p coupling constant α for (a) $q_0 = 10$ and (b) $q_0 = 50$ respectively.

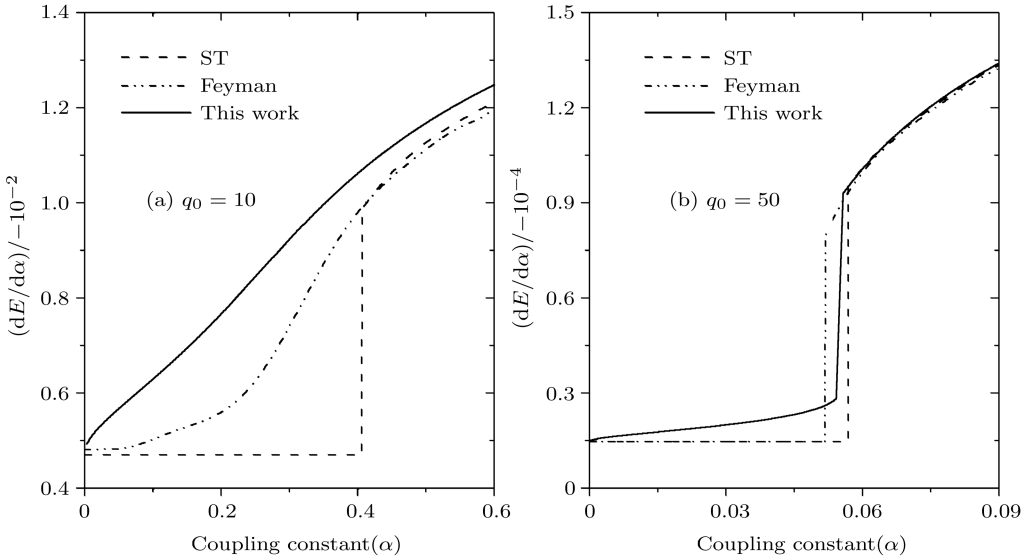


Fig.2. Derivatives of the ground-state energies of the acoustic polarons in the 3D case as functions of the e-p coupling constant α for (a) $q_0 = 10$ and (b) $q_0 = 50$ respectively.

The variational calculations for the ground-state energies of the acoustic polarons in the 3D and 2D cases are numerically performed for different values of q_0, by using expressions (15) and (14) respectively. To compare with the results obtained by using the Feynman path-integral approach,[5,30] we express the energy in units of mc^2 and the phonon vector in units of mc/h in the calculations.

The 3D results for the contributions of the e-LA-p interaction to ground-state energies of the acoustic polarons and the derivatives of the ground-state energies with respect to the e-p coupling constant as functions of α for two values of cutoff wave-vector $q_0 = 10$ and 50 are illustrated in Figs.1 and 2 respectively. For the visual comparison, the results obtained from the Feynman method by Peeters and Devreese and from the Shoji and Tokuda approximation (see Ref.[5]) are also given in the figures.

It is clearly seen in Figs.1 and 2 that the ground-state energy and its derivative vary continuously with the coupling constant α for $q_0 = 10$, but there appears a knee in the derivative of the ground-state energy with respect to α at $\alpha_c \approx 0.26$ (Fig.2(a)) called the "phase transition" critical point, where the polaron state is transformed from quasi-free to self-trapping. As for the case of $q_0 = 50$ the critical point is at $\alpha_c \approx 0.055$, where one can find a knee in the ground-state energy in Fig.1(b) and a discontinuity in the derivative with respect to α in Fig.2(b). It follows that the critical point α_c shifts toward the weaker e-p couplings with increasing the cutoff wave-vector q_0. The calculated results for 3D acoustic polarons in this work are in coincidence with those obtained by using the Feynman approximation in Ref.[5], but we obtain slightly lower ground-state energies.

The numerical results for the ground state energies of the acoustic polarons and their derivatives in the 2D case as functions of the bulk e-LA-p coupling constant α are plotted in Figs.3 and 4 for four values of cutoff wave-vector $q_0 = 10$, 50, 100 and 200.

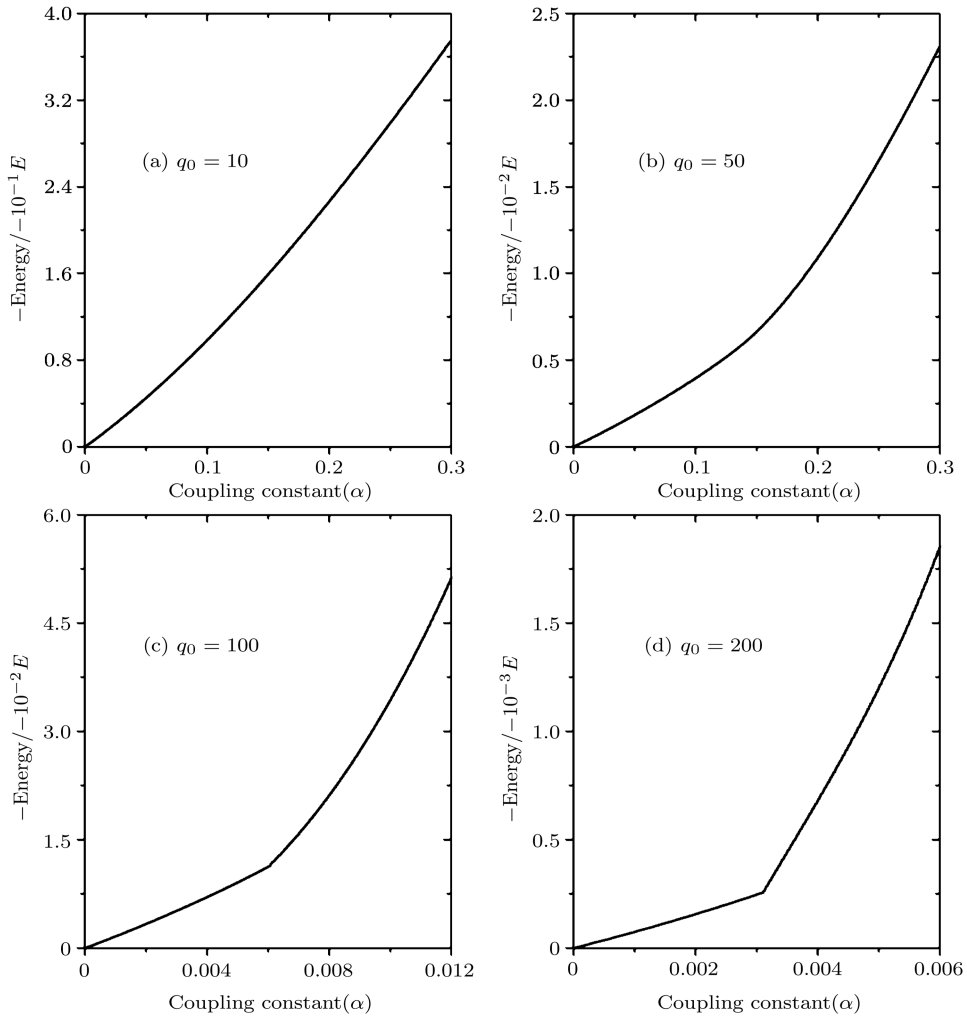


Fig.3. Ground-state energies of acoustic polarons in the 2D case as a function of e-p coupling constant α for $q_0 = 10$ (a), 50 (b), 100 (c) and 200 (d).

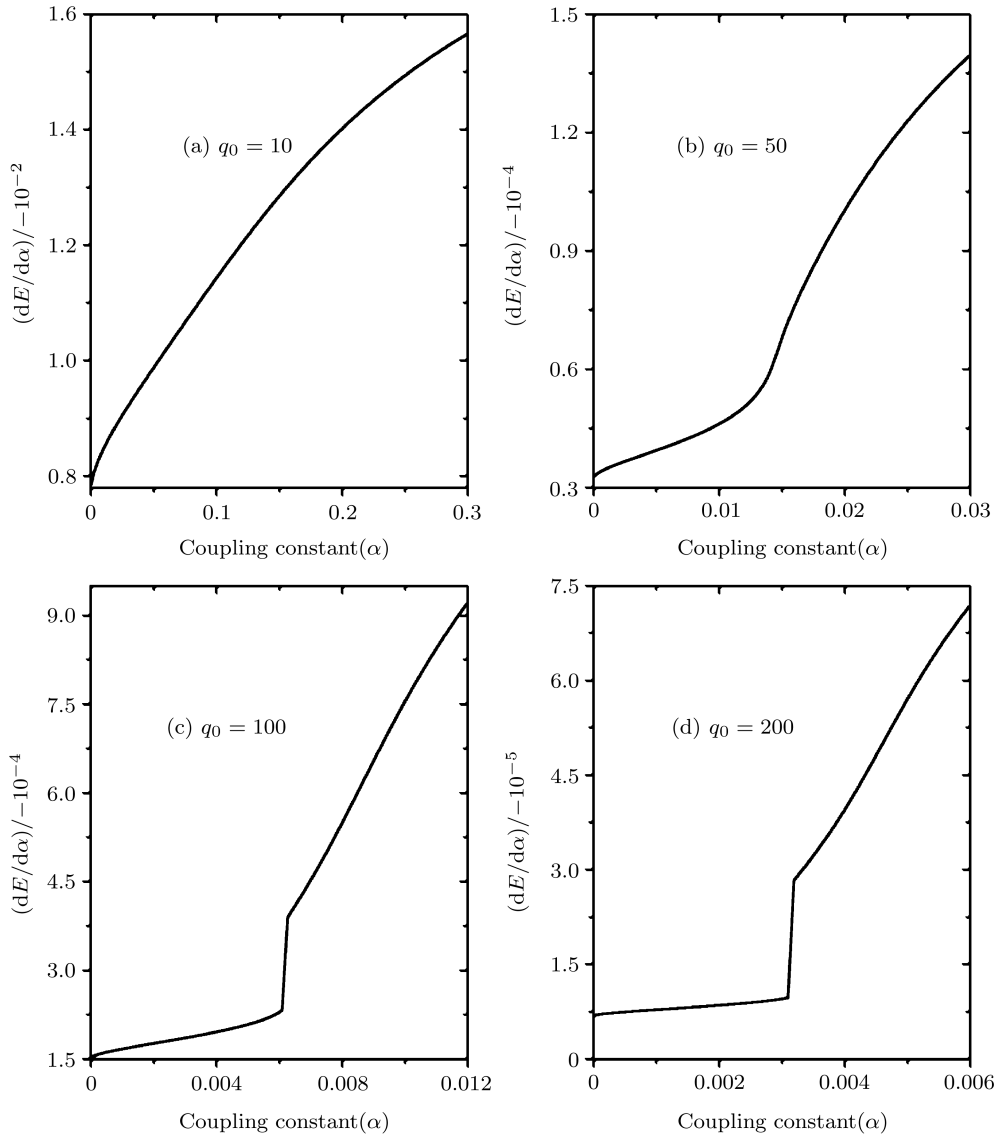


Fig.4. Derivatives of the ground-state energies of acoustic polarons in the 2D case as a function of e-p coupling constant α for $q_0 = 10$ (a), 50 (b), 100 (c) and 200 (d).

Comparing the results in the 2D case with those in the 3D case, one can find that the critical point in the 2D case occurs at a much smaller coupling constant than in the 3D case for the identical cutoff wave-vector because of the stronger e-p coupling effect. For example, for $q_0 = 50$ the critical coupling constant is around 0.055 in the 3D case and 0.012 in the 2D case respectively. Therefore it is concluded that the 2D acoustic polaron is easier to self-trap than the 3D one. It is also found that the position of the critical point is sensitive to the cutoff wave-vector q_0 and shifts toward smaller e-p couplings with increasing the cut-off wave-vector, which is similar to the 3D case. This conclusion is expected and conforms qualitatively to that proposed by Shoji and Tokuda.[4] However, the calculations in Ref.[6] gave quite different results that the obtained 'critical point' α_c in the 2D is higher than that in the 3D case and almost independent of cutoff wave-vector q_0. It seems to indicate that the self trapping of acoustic polarons in the 2D case is more difficult than in the 3D case. The conclusion is unintelligible and contrary to what is given in Ref.[4] and this work. This may be the case that the previous authors used a 3D-like e-p coupling Hamiltonian to describe 2D polarons, and underestimated the e-p effect. The 2D e-LA-p Hamiltonian derived in this paper gives a stronger e-p interaction, which gives a more reasonable conclusion.

It is interesting to check the variation of the critical coupling constant α_c with cutoff wave-vector q_0. The computational results of the first-order derivative of the ground-state energy with respect to α indicate that the 'critical point' in a 2D system is located at $\alpha_c \approx 0.063$ for $q_0 = 10$, 0.012 for $q_0 = 50$, 0.0061 for $q_0 = 100$ and 0.0032 for $q_0 = 200$. It is found that the products $\alpha_c q_0$ of critical coupling constant and cutoff wave-vector for different values of q_0 are almost the same and close to 0.6 (0.63 for $q_0 = 10$, 0.60 for $q_0 = 50$, 0.61 for $q_0 = 100$ and 0.64 for $q_0 = 200$). Therefore one can use $\alpha_c q_0$ as a criterion for judging the presence of the self-trapping transition qualitatively.[3,5] Acoustic polarons in 2D systems can be self-trapping only if αq_0 is larger than $\alpha_c q_0 \approx 0.6$. This feature is also found for 3D systems, where the calculated value of $\alpha_c q_0$ is around 2.6 given by both Ref.[5] and this work. Peeters and Devreese[5] have examined the possibility of the electron self-trapping for some real crystals. They have indicated that the products of αq_0 for 3D semiconductors Ge and Si and III-V compounds as well as alkali halides are too small to lead to the presence of electron self trapping. According to our calculations, the values of critical point α_c of 2D acoustic polarons are all several times smaller than those in the 3D case for the calculated cutoff wave-vectors. It is the case that the value of $\alpha_c q_0$ (around 0.6) in the 2D case is much smaller than that obtained in 3D crystals (around 2.6), so that it is more possible for an acoustic polaron in the 2D case to be self-trapping.

Now we use the theory to judge the possibility of self-trapping transition of acoustic polarons in the 2D and 3D case for real materials according to the criterion of $\alpha_c q_0$.

As pointed out in Ref.[5], the electron self-trapping is impossible to happen to the semiconductors Ge and Si, general III-V compounds and alkali halides.

Recently, much interest has been attracted in the study of the wide band-gap nitride semiconductors, which have become important materials for the fabrication of optoelectronic devices operating in the green, blue, and ultraviolet spectral regions. It is expected to be significant for exploring the features to find whether the electrons in these materials have self-trapping transition. By way of example, we consider GaN and AlN, and the relevant parameters used are listed in Table 1. The bulk e-LA-p coupling constants and the cut-off wave-vectors are $\alpha = 1.09\times10^{-4}$ and 5.94×10^{-4}, $q_0 = 2238$ and 962 for GaN and AlN, respectively. The units of wave-vector q in our calculations are $mc/\hbar = 4.4\times10^6$ and 1.05×10^7 respectively for GaN and AlN.

Table 1. Effective masses of electrons, deformation potentials, lattice constants, mass densities of lattice, transverse elastic constants and sound velocities.

Materials	m/m_e	D/eV	a/Å	$\rho/(\mathrm{kg/m^3})$	$\beta_t/(10^{11}\mathrm{dyn/cm^2})$	$c_t/(\mathrm{m/s})$
GaN	0.19[a]	8.3[b]	3.19[c]	6150[b]	4.42[b]	2681[d]
AlN	0.33[e]	9.5[b]	3.11[c]	3230[b]	4.42[b]	3699[d]

[a]Ref.[31], [b]Ref.[32], [c]Ref.[33], [d]Using $c_t = (\beta_t/\rho)^{1/2}$, [e]Ref.[34].

The calculated product αq_0 for electrons in AlN is 0.57, very close to the critical value 0.6 in 2D systems, so that the self-trapping transition might be possible. As for GaN, even $\alpha q_0 (\approx 0.24)$ can reach a value with the same order of magnitude as that of $\alpha_c q_0$ (0.6), the self-trapping transition is still difficult to observe.

Holes must be easier to self-trap than electrons because of their larger effective masses. For example, the effective masses of the light and heavy-hole are respectively 0.37 and 0.39 in GaN,[34] and the calculated products αq_0 are then 0.91 and 1.01 respectively and both are large enough to have the self-trapping transition in 2D systems, even though it is impossible in the 3D case. Similarly, the light-hole mass in AlN is 0.47[33] and the corresponding product $\alpha q_0 \approx 1.16$ that is smaller than the critical value in the 3D case but larger than that in the 2D case. Therefore the light-holes in AlN can also be self trapping only in the 2D case. However the heavy-hole mass in AlN is 0.73[34] and the product $\alpha q_0 \approx 2.79 (> 2.6)$ that is sufficiently large to have self-trapping either in the 3D or 2D case.

In summary, we have derived a 2D e-LA-p interaction Hamiltonian and used it to investigate the self-trapping transition of 2D acoustic polarons. The calculations give the much stronger e-p coupling contribution to the self-trapping in the 2D case, so that the "phase transition" critical coupling constant α_c is several times smaller than that in 3D systems. It follows that the self-trapping of acoustic polarons is

easier to observe in 2D systems than in 3D systems. The calculations indicate that the holes in GaN and both the electrons and holes in AlN are expected to have the self trapping transition in real 2D systems.

It should be pointed out that we have used a Huybrechts-like variational method, which has been proved available to the whole range of the coupling strength[26,35] by introducing the position-momentum linear combination operators and an extra variational parameter a related to the e-p coupling strength. A comparison of 3D results with those in Ref.[5] still bears out that the variational approach that we used is applicable to seek for the critical point of the self-trapping transition. Nevertheless, due to the fact that the self-trapping electron is captured by the self-inductive potential built by the acoustic phonon field, a bound wave function (such as the Gaussian function)[36] might be better to describe polaron states in the strong coupling limit, which will be considered in the future work.

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