Tr. J. of Physics
23 (1999) , 1021 – 1029.

Confined Bipolarons in the Strong Coupling Limit

R. Tuğrul SENGER, Atilla ERÇELEBİ
Department of Physics, Bilkent University,
06533, Bilkent, Ankara-TURKEY

Received 27.01.1999

Abstract

Within the framework of the strong-coupling polaron theory and the bulk phonon approximation we report the possibility and criteria in achieving stable bipolaron states in confined media. We use a simple model of two electrons constrained within an anisotropic three dimensional parabolic potential box of tunable barrier slopes. Conforming the confining potential from one geometry to the other, we obtain an explicit tracking of the stability criteria as a function of the degree of confinement uncovering all low dimensional major geometric configurations of common interest.

Introduction

Two electrons in an ionic or polar crystal can form a stable bound state, termed a bipolaron, where the surrounding lattice polarization field common to both particles is strong enough to compete with and even dominate over the Coulomb repulsion. Thus, depending on the dielectric properties of the lattice, provided the effective Coulomb repulsion is not larger than a critical strength and the electron phonon interaction is sufficiently strong to overcome that repulsion, there may arise the possibility that a stable bipolaronic state forms [1-12]. The critical conditions which favour bipolaron formation have already been reported in the literature for the bulk and strict two dimensional cases [5-8,11]. Our present concern will be to extend the problem into a more general context which allows us to study the stability criterion as a function of the degree of confinement and hence provide a broader understanding of confined bipolarons through a description encompassing the bulk and quasi-two, -one and -zero dimensional geometries. For this purpose, we introduce a rather simple model of a pair of electrons immersed in the field of bulk LO-phonons and bounded within a harmonic-oscillator potential, i.e., $V_{HO}(\rho,z) = \frac{1}{2}(k_\rho^2\rho^2 + k_z^2 z^2)$. The respective force constants k_ρ and k_z will be treated as adjustable parameters refering to the degree of confinement in the x-y and z directions, respectively. By tuning k_ρ and/or k_z from zero to large values one can obtain an interpolating description of the two-polaron system in various confinement geometries. Such a choice for the confining potential is appealing in the sense that the static depletion fields achieved in microstructures such as quantum wires and dots, which are laterally confined by Schottky gates, exhibit nearly parabolic potentials. Besides this, the usage of quadratic potential profiles greatly facilitates the calculations and leads to concise and tractable analytic expressions.

We should note the fundamental approach followed in this work will be to adopt the so-called *bulk-phonon approximation*, where we take into account solely the generic low dimensional aspect of the dynamical behavior of the confined electrons and take them as interacting with the bulk phonon modes only. We refrain from including any modifications and complications such as those due the contributions from all other kinds of phonon modes, the screening effects, the loss of validity of both the effective mass approximation and the Fröhlich interaction in thin quantum wells and further detailed features. Our approach will primarily be to account for the bulk phonon effects only and provide as simple, yet unifying and comprehesive insight into the confined-bipolaron problem as a function of the effective dimensionality stripped from all other perturbing quantities.

Theory

Using units for which $2m^\star = \hbar = \omega_{LO} = 1$, the Hamiltonian describing the confined electron pair coupled to LO-phonons reads as

$$H = H_e + \sum_Q a_Q^\dagger a_Q + \sum_{j=1,2} \sum_Q V_Q(\alpha_Q e^{i\vec{Q}.\vec{r}_j} + \alpha_Q^+ e^{-i\vec{Q}.\vec{r}_j}), \tag{1}$$

$$H_e = \sum_{j=1,2} p_j^2 + \sum_{j=1,2} \frac{1}{4}(\Omega_\rho^2 \rho_j^2 + \Omega_z^2 z_j^2) + \frac{U}{|\vec{r}_1 - \vec{r}_2|}, \tag{2}$$

where

$$\Omega_\rho = \sqrt{k_\rho/m^\star \omega_{LO}^2}, \text{ and } \Omega_z = \sqrt{k_z/m^\star \omega_{LO}^2}. \tag{3}$$

In the above, a_Q and $a_Q^\dagger$ denote the phonon operators, and $\vec{r}_j = (\vec{\rho}_j, z_j), (j = 1,2)$, are the positions of the electrons in cylindrical coordinates. Similarly, p_j refer to the corresponding momenta. The interaction amplitude is related to the phonon wavevector $\vec{Q} = (\vec{q}, q_z)$ through $V_Q = (4\pi\alpha)^{1/2}/Q$. The coupling constant is given, in terms of the high frequency and static dielectric constants of the material, by

$$\alpha = \frac{e^2}{2}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0}\right), \tag{4}$$

in terms of which the unscreened Coulomb repulsive amplitude is

$$U = e^2/\epsilon_\infty = 2\alpha/(1-\eta), \tag{5}$$

where $\eta = \epsilon_\infty/\epsilon_0 < 1$.

Tuning the dimensionless frequencies Ω_ρ and/or Ω_z from zero to large values one achieves a broad display of the phase stability of the bipolaron state as a function of the effective dimensionality extending from the bulk to the extreme limit of strict two dimensions and interpolating further over a wide range of all major geometric configurations. Setting either $\Omega_\rho = 0$, or else, $\Omega_z = 0$, and fixing the remaining parameter at non-zero finite values, the geometry conforms respectively to the quasi-two dimensional (Q2D) slab-like structure or the quasi-one dimensional (Q1D) "free-standing-wire" configuration. Hereafter, whereever necessary to facilitate the notation, we shall use Ω to mean $\Omega_\rho(\Omega_z)$ when $\Omega_z(\Omega_\rho) = 0$. In the spherically symmetric box-type configuration we shall simply set $\Omega_\rho = \Omega_z = \Omega$.

The approximation that we adopt here is the conventional Pekar adiabatic theory [13] which imposes a product Ansatz separable in the particle and phonon coordinates, i.e.,

$$\Psi = \Phi(\vec{R}, \vec{r}) \times \exp \sum_Q g_Q (a_Q - a_Q^\dagger)|0\rangle \tag{6}$$

In the above, the exponential operator acting on the phonon vacuum is the displaced oscillator transformation, where g_Q is a variational parameter determined from the requirement that $\partial\langle\Psi|H|\Psi\rangle/\partial g_Q = 0$, yielding

$$g_Q = V_Q \sum_{j=1,2} \langle\Phi|e^{\pm i\vec{Q}.\vec{r}_j}|\Phi\rangle. \tag{7}$$

For the particle part of the trial state (6), we assume variational oscillator-type wavefunctions separable in the centre of mass, $\vec{R} = (\vec{r}_1 + \vec{r}_2)/2$, and the relative coordinates $\vec{r} = \vec{r}_1 - \vec{r}_2$, i.e.,

$$\Phi(\vec{R}, \vec{r}) = N\sqrt{r_\rho^2 + r_z^2} \exp\left\{-\frac{1}{2}\lambda_1^2(R_\rho^2 + \mu_1^2 R_z^2)\right\} \times \exp\left\{-\frac{1}{2}\lambda_2^2(r_\rho^2 + \mu_2^2 r_z^2)\right\}, \tag{8}$$

in which $\vec{R}_\rho$ and R_z stand for the lateral and z components of the center of mass position vector, and the components $\vec{r}_\rho$ and r_z have similar meanings for the relative position vector. N is the normalization constant, and the factor $r = \sqrt{r_\rho^2 + r_z^2}$ multiplying the exponentials ensures that $\Psi|_{r=0} = 0$; hence the electrons are repulsively kept separated.

The bipolaron ground state energy can be obtained through a numerical minimization of $E_g = \langle\Psi|H|\Psi\rangle$ with respect to the set of four variational parameters: $\{\lambda_i, \mu_i\}, (i = 1, 2)$. In our numerical calculations we shall trace the domain of stability of the bipolaron as a function of α and the confining parameters Ω_ρ and Ω_z.

Results and Conclusions

The criterion for which a stable bipolaronic state can form is that the energy of the system of two interacting polarons should be lower than twice the one-polaron energy.

Stating alternatively, for the bipolaron formation to be favorable, one should have

$$E_g < 2E_g^{(1)}, \tag{9}$$

where $E_g^{(1)}$ refers to the one-polaron ground state energy, calculated within an identical framework to the bipolaron system. For this purpose, we shall take the one-polaron energy results from a previous paper [14] devoted to the study of the one-polaron problem within exactly the same context as in the present consideration.

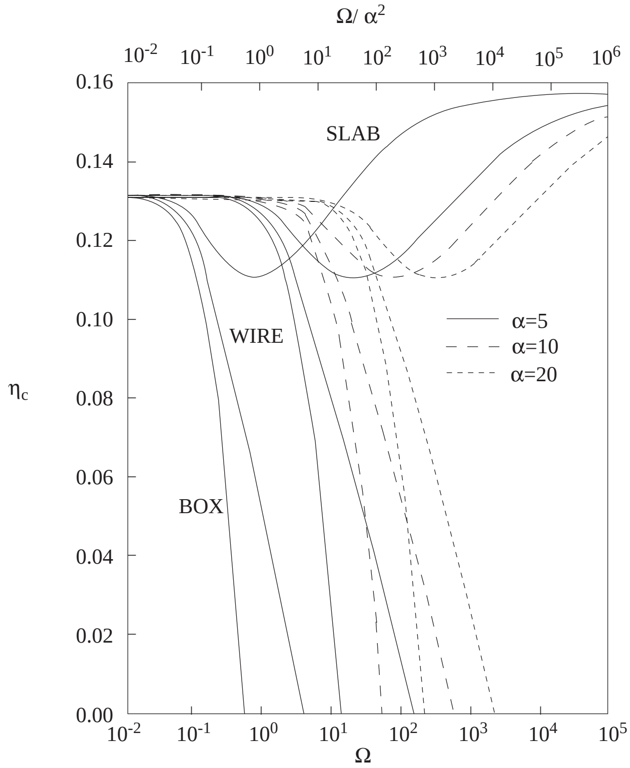

Figure 1. The critical ratio η_c as a function of the degree of confinement for the quasi-two, -one and -zero dimensional configurations displayed respectively by the upper, middle and lower curves in each of the four similar sets of plots, the three of which are in thin font and one in thick. The thin curves display the variation of η_c against bare Ω (the bottom axis) for $\alpha = 5, 10$ and 20, plotted respectively in solid, dashed and dotted lines. The assembly of thick solid curves plotted against Ω/α^2 (the top axis) is a universal duplicate of the concent of the thin curves regardless of the value of α.

Of immediate relevance to this work are the recent analyses of the problem in strict two dimensions [5-8,11], where it is observed that the critical Coulomb repulsion coefficient, below which bipolaron formation is favourable, takes on a larger value than in three dimensions. Specifically, within the same approximate adiabatic theory and the same oscillator-type particle wavefunction separable in the centre of mass and relative coordinates, the critical η which governs the domain of stability ($\eta < \eta_c$) is found to be 0.131 in the bulk (3D), and 0.158 in two dimensions (2D), respectively [7]. In the same reference, it has also been noted that, since in the strong-coupling the energies are proportional to α^2, there is no critical value of α in this scheme. The finding that there is no relation to a critical α in bulk and two dimensions is an artifact of the strong-coupling theory rather than any intrinsic property of the two-polaron system or the Fröhlich Hamiltonian.

Before we present our results, we would like to mention that, if the energies are scaled by α^2 and lengths by α, i.e., $E \rightarrow E\alpha^2$ and $L \rightarrow L\alpha$, the only modification in the Hamilltonian, Eqs. (2-3), would be to replace the confining parameter Ω by Ω/α^2 and the Coulomb coefficient U by U/α. Thus, in a representation where the critical value of parameter $\eta(= 1 - 2/(U/\alpha))$ is to be plotted against Ω/α^2, rather than Ω, we find that irrespective of the value of α one can display the phase boundary on a single universal curve for each individual geometry. In every case, whatever the geometric configuration is, the ground state energy is seen to be proportional to the square of the coupling constant, i.e. $E_g = -C\alpha^2$, where C, the corresponding coefficient of proportionality, bears a functional relation solely to Ω/α^2 for both the single-polaron and two-polaron systems. Therefore, in the foregoing particular plot for which the abscissa is expressed in units of the ratio Ω/α^2, one can conveniently assign α any arbitrary large value with no loss in generality.

In Fig. 1 we provide a series of plots portraying the variation of the critical value η_c as a function of the degree of confinement for the quasi-two, -one and -zero dimensional configurations. The computer runs performed for three different coupling constants ($\alpha = 5, 10$ and 20) are plotted as thin curves with the abscissa taken as bare Ω. Conforming the scale of the abscissa from Ω to Ω/α^2, we find out that, whatever the value of α, the thin curves coalesce together and universally map onto one single curve as displayed in thick solid font in the figure.

Holding α fixed at any desired value and following the variations in η_c as Ω is turned on, we first note that for each of the aforementioned dimensionalities the critical η, starting from the common 3D-value, $\eta_c^{(3D)} = 0.131$, displays in common a decreasing trend where the respective decay rates are observed to become faster as the effective dimensionality is reduced from two to one, and to zero. Thus, in the wire-and box-type configurations the critical Coulomb strength below which a bipolaron forms lies deviated considerably below the corresponding value in the slab-type geometry. We therefore see explicitly that lower the dimensionality, the more unlikely it is to realize the bipolaron state to form; and in particular, in the box-type confinement, even a small value of η will be sufficient enough to lead the Coulomb force to oppose and dominate over the phonon-coupling-induced localization of the composite assembly of the two nearby electrons (cf., Ref.[11]). Clearly, with increasing degree of confinement (i.e., with growing Ω_ρ and/or Ω_z), the Coulomb repulsion is steadily strengthened as the particles are squeezed to get closer; and one expects the rate at which this happens be most prominent in the QOD-configuration where the electrons are pushed towards one another from all radially inward directions. In the Q1D- and Q2D-configurations, however, the electrons are free to expand and relax themselves respectively in either one or two directions; thus resulting with comparatively weaker Coulomb repulsion and weaker dominating strength over the lattice polarization field holding the particles together.

We should note that in confined systems it is not only the Coulomb repulsion strength which gets pronounced, but also the phonon-coupling becomes pseudo-enhanced leading to a more effective and deeper polaronic binding to oppose and counterbalance the repulsive forces. The competitive interrelation between these aspects of the problem determines the phase boundary and, moreover, may even pose a salient feature stemming from a cross-over of this competition as the confining parameters are varied. In particular, in the Q2D slab-geometry where this feature shows up most prominently, we observe that the critical η does not display a monotonically increasing behavior interpolating between the 3D- and 2D- limits, but instead reaches its two dimensional value [7] ($\eta_c^{(2D)} = 0.158$) only after having passed through a minimum; thus reflecting an explicit image of the dominating effect of either the Coulomb repulsion or the phonon mediated attraction over the other.

For a complete description covering the ranges between all possible extremes of the effective dimensionality, we set $\alpha = 10$ and construct the phase-picture of the bipolaron system over reasonably broad ranges of Ω_ρ and Ω_z (cf., Fig. 2). Starting from the left corner at the top we see that η_c maintains its 3D-value ($\sim$0.131) until the polarons start to feel the boundary potential, and beyond the flat plateau thus formed, the size effects start to influence the Coulomb and electron-phonon interactions and alter the bipolaron stability greatly. On following the directions along the Ω_ρ- and Ω_z-axes and the line $\Omega_\rho = \Omega_z$, we achieve the phase boundary for the quasi-one, -two and -zero dimensional confinements, respectively. Clearly, the deep valley lying in between the 3D, annd 2D-plateaus and downhill between the flanks provides a broader large scale portray of the aforementioned feature pertaining to the crossover of the dominating strengths of the repulsive and attractive forces induced by the Coulomb and electron-phonon interactions.

A further distinctive feature of the problem intruded by the confining potential is that, except in the extreme limits, $\Omega_\rho = 0$ and $\Omega_z = 0$ or ∞, the phase boundary is seen to be sensitive to the coupling constant. Clearly, in Fig. 1 we note that the set of all curves drawn for different α originate from the common 3D-value, but however display a succession of translationally displaced profiles as α is varied. It should be noted that the place at which the curves start to deviate below the 3D-value, shifts towards larger Ω for stronger phonon coupling, since for large α the polarons are already in a highly localized configuration and a small-sized bipolaron becomes influenced by the confining boundary only for large Ω. Alternatively stating, for large α one requires inevitably larger values of Ω for the geometric confinement to take and dominate over the further confinement induced by phonon coupling. Peculiar to the slab geometry, we also observe that in the limit $\Omega \to \infty$, regardless of α, all plots merge asymptotically to the same

2D-value. This observation is, in fact, totally consistent with the aforementioned remark that, in the bulk and two dimensional cases, there is no critical value for α. However, for configurations other than the integer dimensional space limits, the geometric confinement is seen to affect the phonon-coupling induced localization of the system; thus resulting in an explicit relevance of the coupling constant to the formation of bipolarons in confined media. ln all the three basic geometries in Fig. 1 we observe that, for a given non-zero and finite Ω, larger the coupling constant, greater should be the Coulomb strength to violate the stability of the bipolaronic state and set the polarons apart from one another. In the Q2D-configuration with Ω selected as 10, for instance, we calculate the critical Coulomb coefficient as $U_c = 7.97, 16.12$ and 32.52 for $\alpha = 5, 10$ and 20, respectively. Apparently, the same is true for the Q1D- and Q0D-configurations where for $\Omega = 10$ we correspondingly obtain $U_c = 7.69(7.31), 15.96(15.79)$ and 32.48 (32.44) in the wire (box) geometry.

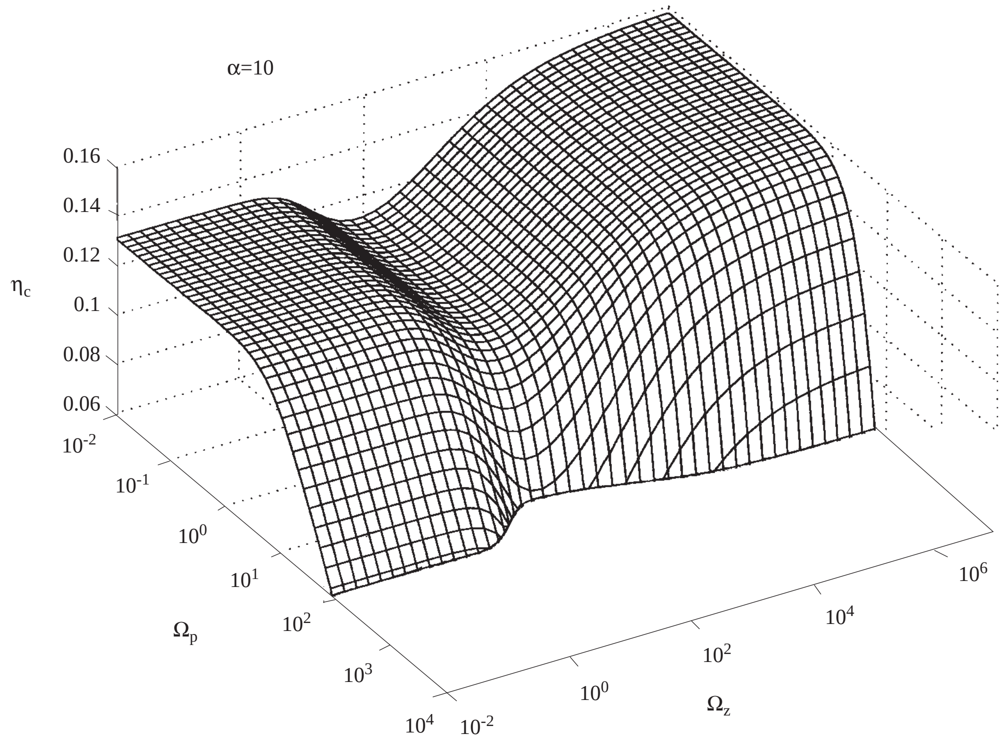

Figure 2. The critical ratio η_c as a function of the confining parameters for $\alpha = 10$.

To give somewhat more impact to the role which parameter Ω plays in making α show up explicitly in the phase picture, we repeat our numerical runs for a display against α and provide an alternate presentation of the content of Fig. 1. An immediate glance at the set of curves plotted in Fig. 3 reveals that, except in the extreme limits of small and very large

Ω, the critical η displays rather drastic variations as α is varied. However, as the limit $\Omega \to 0$ is approached (cf., the curve for $\Omega = 1$), we clearly observe that η_c tends to become independent of α and simply take on the bulk value (0.131). Examining the variation of η_c against α over a number of distinctive Ω values, one can easily trace out the means η_c gradually loosing its dependence on α and conforming to the constant bulk limit as Ω is made smaller and eventually turned off. Similarly, in the slab-type configuration (cf., Fig.3-a), referring to the set of curves plotted for considerably high degrees of confinement, $\Omega >> 1$, we see that as the strict two dimensional limit is attained, the relevant curves tend to approach to the horizontal straight line $\eta_c = 0.158$, being stripped down to its bare two dimensional characterization with no α-dependence at all.

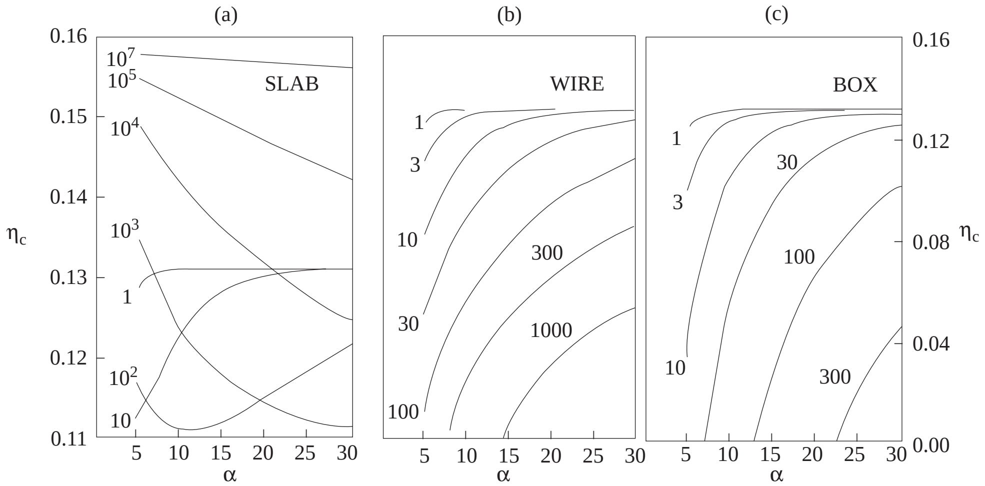

Figure 3. The critical ratio η_c as a function of α for a series of different Ω values in (a) the quasi-two, (b)-one, and (c)-zero dimensional configurations.

As one further comment, we should note that in the slab configuration, whatever the values of Ω and α are, there is always a non-zero critical η below which a stable bipolaronic state can be realized. In the wire- and box-type confinements however, no matter how weak the Coulomb strength might be set, there is always an upper value for Ω beyond which the bipolaron breaks up into two individual polarons. Increased values of α can only support the bipolaron to converse its stability at correspondingly higher degrees of confinement.

In this article we have studied the possibility and criteria in achieving stable bipolarons in low dimensionally confined media. The "deformable potential box"-model adopted in this work allows us to attain a simple and yet comprehensive review of the two-polaron problem within a unifying scheme interpolating between the bulk and all low dimensional geometric configurations of general interest. It has been illustrated that in structures with reduced dimensionality, the phase description displays an explicit relevance to the phonon-coupling parameter α, distinguished from that reported earlier for the strongly coupled bipolaron in three and two dimensions. It has been shown further that in conforming the potential box to a thin quantum well, the the critical ratio of the dielectric constants may undergo an interesting variation, exhibiting a decrease first, then ascending and eventually going over to its topmost two dimensional value.

Acknowledgements

One of the authors (R.T.S.) acknowledges the partial support of TÜBİTAK-Münir Birsel Vakfı

References

[1] H. Hiramoto and Y. Toyozawa, *J. Phys. Soc. Jpn.* **54** (1985) 245

[2] J. Adamowski, *Phys. Rev.*B **39** (1989) 3649.

[3] T. K. Mitra, 1989 *Phys. Lett.* A **142** (1989) 398.

[4] F. Bassani, M. Geddo, G. Iadonisi and D. Ninno *Phys. Rev.* B **43** (1991) 5296

[5] G. Verbist, F. M. Peeters and J. T. Devreese *Phys. Rev.* B **43** (1991) 2712.

[6] S. Sil, A. G. Giri and A. Chatterjee *Phys. Rev.*B **43** (1991) 12642

[7] G. Verbist, M. A. Smondryrev, F. M. Peeters and J. T. Devreese *Phys. Rev.*B **45** (1992) 5262

[8] J. Adamowski and S. Bednarek *J. Phys.: Condens.Matter* **4** (1992) 2845

[9] A. Chatterjee and S. Sil *Int.J.Mod.Phys.*B **7** (1993) 4763

[10] F. Luczak, F. Brosens and J. T. Devreese *Phys. Rev.*B **52** (1995) 12743

[11] S. Sahoo, *J.Phys.: Condens. Matter* **7** (1995) 4457

[12] S. Mukhopadhyay and A. Chatterjee *J.Phys.: Condens. Matter* **8** (1996) 4017

[13] S. I. Pekar, Untersuchungen über die Elektronentheorie der Kristalle (1954) Berlin, Akademie

[14] T. Yıldırım and A. Erçelebi J. Phys.: Condens. Matter **3** (1991) 1271