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Polaron mass of carriers in a thin film on ionic substrates

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A new approach to establishing a strong electron–phonon interaction in heterostructures is proposed. A three-layer structure consisting of an ionic substrate, a semiconductor film, and a covering dielectric (with air or vacuum possibly serving as such a dielectric) is examined. Interface optical phonons emerge near the heterointerface. Their parameters are governed by the dielectric properties of the substrate. It is demonstrated that the effective mass of carriers in the film is altered in the presence of interface phonons. Depending on the substrate ionicity, the magnitude of this change may vary from several tens to hundreds of percent. It is shown that the conditions for strong electron–phonon interaction may be established in a large number of semiconductor films. Measurements of the effective mass of carriers in identical films positioned on different substrates should make it possible to identify a transition from a weak electron–phonon interaction to a strong one.

Keywords: electron–phonon interaction, effective mass, interface phonons, polaron, thin films.

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1. Introduction

A significant progress in the characterization of phonon properties of nanostructures has been achieved in recent years [1,2]. It was found that the presence of interface optical phonons needs to be taken into account in order to characterize accurately the thermal, optical, and transport properties of such objects. Interface phonons take on particular importance in the examination of the electron–phonon interaction [3]. The interaction of charged particles with interface optical phonons is the mechanism that may lead to the formation of large-radius quasi-two-dimensional polarons. The interaction magnitude may vary severalfold even in structures of one and the same type [4]. This makes it theoretically possible to implement a transition from a weak electron–phonon interaction to a strong one in nanostructures.

A slightly different implementation of a transition from a strong electron–phonon interaction to a weak one is proposed in the present study. Such a transition may be identified by depositing a thin film of one and the same material on a series of different substrates with a high ionicity. Interface phonons emerging at the substrate/film interface should induce polarization, which is governed by the dielectric properties of the substrate, in the film. This allows one to examine the manifestation of polaron effects of a varying magnitude in identical films.

2. Polaron mass

The variation of effective carrier mass in a film is regarded in the present study as a manifestation of the electron–phonon interaction. The theory of this polaron mass in two-dimensional and quasi-two-dimensional objects is well-developed. In the case of a weak or moderate interaction, the polaron mass is given by [5]

$$m_{pol} = m^*\left(1 + \frac{\pi}{8}\alpha\right), \quad \alpha < 1, \tag{1}$$

where m^* is the effective carrier mass in a film that is not in contact with ionic materials; α is a dimensionless parameters of the electron–phonon interaction. Note that the value of this parameter in the considered system depends on the dielectric properties of the substrate. In the case of a strong interaction with $\alpha \gg 1$, the polaron mass is given by [5]

$$m_{pol} = 0.733\alpha^4 m^*. \tag{2}$$

It follows from the comparison of formulae (1) and (2) that the formal transition from a strong interaction to a weak one occurs at $\alpha \approx 1.19$. It will be demonstrated below that both strong and weak electron–phonon interactions may be established in one and the same film with use of different substrates.

3. Parameters of the electron–phonon interaction

Let us determine the value of parameter α in the dielectric continuum model [6]. This model provides a fairly accurate description of the actual interaction in a film if polaron state radius r_{pol} is greater than lattice constant a_0:

$$r_{pol} > a_0, \tag{3}$$

where $r_{pol} = \sqrt{\hbar/\alpha m^* \omega_{\text{int}}}$, and ω_{int} is the frequency of interface optical phonons. Condition (3) is satisfied for all the compounds discussed below. Let us consider a three-layer structure consisting of a substrate, a film, and a coating. This coating may take the form of vacuum, air, or a nonpolar dielectric with constant permittivity ε. The substrate and the film have permittivities $\varepsilon_s(\omega)$ and $\varepsilon_f(\omega)$, respectively. It is assumed hereinafter that the ionicity of the substrate is much higher than the ionicity of the film. This is the case when the substrate is central to the emergence of polaron states. We assume that frequency dependences $\varepsilon_i(\omega)$ for both the substrate and the film take the form

$$\varepsilon_i(\omega) = \varepsilon_{\infty,i}\,\frac{\omega^2 - \omega^2_{\mathrm{LO},i}}{\omega^2 - \omega^2_{\mathrm{TO},i}}. \tag{4}$$

Let us determine the spectrum of interface phonons in the considered structure in accordance with [4,6]. It is defined by the solution of the following equation:

$$e^{-qa}\,\frac{\varepsilon_f(\omega) - \varepsilon_s(\omega)}{\varepsilon_f(\omega) + \varepsilon_s(\omega)} = e^{qa}\,\frac{\varepsilon_f(\omega) + \varepsilon}{\varepsilon_f(\omega) - \varepsilon}, \tag{5}$$

where a is the film width and q is the magnitude of the two-dimensional phonon vector in the film plane. The solution of Eq. (5) specifies two types of interface phonons. The first one features two waves with their intensity maximum positioned at the film/substrate interface. The second type of interface phonons features only one wave with its intensity maximum lying between the film and the surface layer.

It will be demonstrated below that the strongest electron–phonon interaction corresponds to one of the waves of the first type, which is close in its parameters to LO phonons in the substrate. It should be noted that the parameters of this excitation remain almost unchanged in the limit of small q values, where

$$qa \ll 1. \tag{6}$$

The typical q values are on the order of the reciprocal radius of the polaron state:

$$qr_{pol} \approx 1. \tag{7}$$

Therefore, condition (6) specifies an upper bound for the film thickness. The polaron radius in the structures considered below falls within the range from several nanometers to several tens of nanometers. The strongest electron–phonon interaction is established when the film thickness does not exceed the polaron radius. The influence of interface optical phonons on the effective carrier mass variation in thicker films is significantly weaker. If inequality (6) is satisfied, Eq. (5) becomes considerably simpler, giving rise to condition

$$\varepsilon_s(\omega) = -\varepsilon, \tag{8}$$

which formally corresponds to the equation for the spectrum of interface phonons at a single heterointerface between an ionic substrate and a nonpolar dielectric. The field of these phonons penetrates into the film and interacts with charged particles localized there. The frequencies of interface phonons of this type are

$$\omega^2_{\mathrm{int}} = \frac{\varepsilon_{s,\infty}\omega^2_{\mathrm{LO},s} - \omega^2_{\mathrm{TO}}\varepsilon}{\varepsilon_{s,\infty} - \varepsilon}. \tag{9}$$

Expression (9) characterizes actual interface phonons only if $\varepsilon_{s,\infty} > \varepsilon$. Their frequency lies between the frequencies of LO and TO phonons of the substrate; i.e.,

$$\omega_{\mathrm{TO},s} < \omega_{\mathrm{int}} < \omega_{\mathrm{LO},s}. \tag{10}$$

If the dependence of frequencies of interface phonons on the wave vector is taken into account (in the case when an exact Eq. (5) is used), small corrections, which normally do not violate condition (10), emerge.

In addition to the solution for interface phonons (9), the system also retains intrinsic LO and TO thin-film modes, which evolve into two other interfacial modes in accordance with Eq. (5) at finite values of two-dimensional vector $\mathbf{q}$. However, the interaction of charged particles with films of a weakly ionic compound is considerably weaker. The Hamiltonian of interaction with charged particles for an interfacial mode may be presented in the standard form [3]:

$$H_{\mathrm{int}} = \sum_{\mathbf{q}} \left(\frac{2\lambda\omega_{\mathrm{int}}}{L^2}\right)^{1/2} \frac{\exp(i\mathbf{q}\rho)}{\sqrt{2q}}\, f_{\mathrm{int}}(q)\left[a_{\mathrm{int}}(\mathbf{q}) + a^{+}_{\mathrm{int}}(\mathbf{q})\right], \tag{11}$$

where L^2 is the normalizing film area and $a_{\mathrm{int}}(\mathbf{q})$ and $a^{+}_{\mathrm{int}}(\mathbf{q})$ are the creation and annihilation operators for the studied mode. If condition (6) is satisfied, factor $f_{\mathrm{int}}(0)$ assumes a rather simple analytical form. It differs from the corresponding factor in a quantum well with symmetric ionic barriers only in the fact that the common expression for optical permittivity

$$1/\varepsilon_{opt} = 1/\varepsilon_\infty - 1/\varepsilon_0$$

is substituted in it by modified quantity $\varepsilon^{\mathrm{int}}_{opt}$

$$\varepsilon^{\mathrm{int}}_{opt} = \varepsilon_{opt}\,\frac{\varepsilon_{0s}\varepsilon_{\infty s}}{2(\varepsilon_{0s} - \varepsilon)(\varepsilon_{\infty s} - \varepsilon)}, \tag{12}$$

which depends on the permittivity of the material under the film. Quantity $\varepsilon^{\mathrm{int}}_{opt}$ from (12) is the one that is found in all parameters characterizing the electron–phonon interaction in our system (specifically, dimensionless interaction parameter α_{int}, which is analogous to the Fröhlich constant for bulk materials). In accordance with (11), (12), the expression for α_{int} may be written as

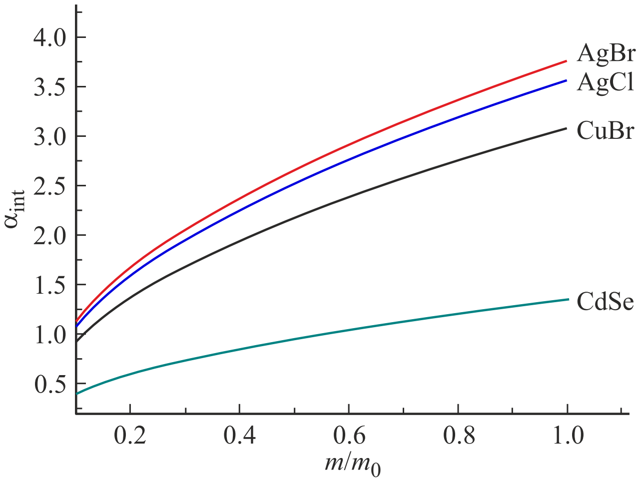
$$\alpha_{\mathrm{int}} = \frac{e^2\sqrt{m\omega_{\mathrm{int}}}}{2\omega_{\mathrm{int}}\varepsilon^{\mathrm{int}}_{opt}\hbar^{3/2}}. \tag{13}$$

This quantity α_{int} defines the polaron carrier mass in Eqs. (1) and (2). The values of effective interaction parameter α_{int} and the polaron electron mass for several semiconductor films on substrates made from ionic compounds are listed

Values of electron–phonon interaction parameter α_{int} and polaron mass m_{pol},which was calculated using formulae (1) or (2), for different films on AgBr, AgCl, and CuBr substrates

Substrate	AgBr				AgCl				CuBr			
Film	α_{int}	m	m_{pol} (1)	m_{pol} (2)	α_{int}	m	m_{pol} (1)	m_{pol} (2)	α_{int}	m	m_{pol} (1)	m_{pol} (2)
TGLs	0.68	0.033	0.04	–	0.65	0.033	0.04	–	0.56	0.033	0.04	–
GaAs	0.94	0.063	0.09	–	0.9	0.063	0.09	–	0.78	0.063	0.08	–
CdSe	1.46	0.15	–	0.49	1.38	0.15	–	0.4	1.2	0.15	–	0.23
GaN	1.68	0.2	–	1.17	1.6	0.2	–	0.95	1.38	0.2	–	0.53

Note. The values of electron mass m for film materials were taken from [7]. All masses are given in units of free electron mass m_0.



Dependence of the electron–phonon interaction parameter on electron mass m/m_0 in the film.

in the table. The parameters needed for calculation are given in [7,8]. Formulae (1) or (2) were used in calculation depending on the value of parameter α_{int}. It can be seen from the table that the electron–phonon interaction with parameter α_{int} being close to unity may theoretically be established even in a GaAs film. The high ionicity of the substrate allows one to implement a transition to the strong electron–phonon interaction regime in films where the effective mass of carriers exceeds $0.1m_0$ (m_0 is the free electron mass) in the free-standing state. It was assumed in calculations that the permittivity of the material above the film surface is $\varepsilon = 1$. The interaction gets weaker at higher values, and is becomes harder to implement the strong coupling regime. The parameters for ultrathin films of two-layer graphene (TGLs) are also shown in the table as an example.

Since the dielectric properties of the film are insignificant in the studied system, our approach is theoretically applicable to graphene and other films of extreme two-dimensionality. The carrier mass in two-layer graphene may be increased by several tens of percent. At the same time, the effective mass for dichalcogenides of transition metals on ionic substrates is expected to increase severalfold, and the strong interaction regime should be established. However, since the carrier mass in a free-standing film is large in this case [9,10], condition (3) may be violated at a high substrate ionicity. Therefore, numerical values of the polaron mass calculated using (1) and (2) are not quite correct. That said, the qualitative effect of a strong variation of the carrier mass with different substrates should persist. This effect is the likely reason behind the considerable discrepancy between calculated and experimental values of the effective carrier mass in dichalcogenides, which was noted in [9,10]. However, further studies, the results of which may be reported in a separate paper, are needed to obtain quantitative estimates of the polaron mass.

The magnitude of polaron effects depends to a considerable extent on the carrier mass in a free-standing film. This dependence is presented in the figure, which demonstrates that a strong interaction emerges in most semiconductor films if a substrate is made from $\mathrm{A^{I}B^{VII}}$ materials. Using a substrate fabricated based on $\mathrm{A^{II}B^{VI}}$ compounds, one may observe a significant enhancement of the carrier mass in a film, but the strong interaction regime is unlikely to be established.

The results obtained in the present study correspond to substrate materials with a cubic symmetry. If less symmetric materials are used as substrates, the considered interaction becomes weaker in most cases. However, this issue warrants detailed investigation. For example, an insignificant and an almost complete suppression of the interaction of charged particles with interface optical phonons are both possible in symmetric quantum wells with barriers made from uniaxial materials: the magnitude of suppression depends on the degree of anisotropy of their dielectric parameters [11].

4. Conclusion

Thus, a considerable enhancement of the effective carrier mass is to be expected in thin semiconductor films on ionic substrates. The magnitude of this effect depends on the substrate ionicity and the effective carrier mass in a free-standing film. The effective mass on substrates made from $\mathrm{A^{II}B^{VI}}$ compounds should change by several tens of percent. When $\mathrm{A^{I}B^{VII}}$ compounds are used as a substrate, the effective carrier mass in a film may increase severalfold. The strong electron–phonon interaction regime may be established in this case. The considered effects should be taken into account in the examination of relaxation processes of hot carriers and various optical and transport properties of structures similar to the ones discussed above. An experimental verification of the obtained results should help estimate the correctness of theoretical models characterizing the electron–phonon interaction.

Conflict of interest

The authors declare that they have no conflict of interest.

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