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Criteria for the appearance of a periodical component in the polaron distribution function

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In this paper we establish criteria of instability of the homogeneous large polaron gas versus the periodic state. We study, in the framework of the Bogolyubov approach of the equilibrium distribution functions, how this transition depends on the parameters of the polar medium (dielectric parameters, electron-phonon coupling constant, polaron density). The temperature dependence of the transition is analyzed, indicating that the stability of the periodic phase can be achieved in a given range of temperatures, depending on the material and the polaron density. The comparison of the total energies of the periodic and homogeneous phases does confirm that the stability of the periodic phase corresponds to the lowest-energy configuration.

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I. INTRODUCTION

The strong electron-phonon coupling and polaronic effects play an important role in determining the properties of several compounds, such as high-temperature cuprate superconductors,[1] colossal magnetoresistence manganites,[2] nickelates and quasi-one-dimensional materials (for example, organic conjugated polymers[3]).

A problem of large interest is that of the instability of the polaron gas with and without the interaction with an external field. Many specific physical situations and models have been considered to show the crossover from the simple homogeneous polaron gas toward structured phases. Due to the complexity of the problem, a general theory of such instabilities is still missing.

Starting from the studies of Holstein on a single-polaron Hamiltonian,[4] it has been shown that the ground state can contain the features of both the large and the small polaron.[5] The coexistence of both the large and the small polaron together with the double- and superexchange magnetic effects, has been shown to be significant in manganites, such as $La_{1-x}Ca_xMnO_3$. Near the metal-insulator transition induced by the temperature or hole doping, the system segregates in antiferromagnetic or paramagnetic small-polaron and ferromagnetic large-polaron domains having different lattice distortions. This leads to strong modifications on the infrared absorption, also in presence of an external magnetic field or isotopic oxygen substitution.[6]

The quantum lattice fluctuations in the ground state of a half-filled electron band are the origin of the Peierls instability[7] of the one-dimensional molecular-crystal model both with spin-less and with spin-1/2 electrons. In the spinless case an order-disorder transition is found on increasing the phonon frequency. On the contrary, the system with spin-1/2 electrons is always ordered, but the order parameter is a decreasing function of the phonon frequency. At high phonon energies, the charge density wave ordering prevails and in the limit of infinite phonon energy the system becomes borderline between charge-density ordering and superconducting state. The signature of such fluctuations can be found in the infrared conductivity.[8] Finally, in cuprates superconductors the formation of different polaron phases (self-trapped or not) or of stripes has been largely studied in the attempt of finding a reliable mechanism for high-T_c superconductivity.[9]

The instabilities of a large polaron system have been studied in the past. For example, the formation and the melting of the polaron Wigner crystal has been considered. It has been found that the polaron Wigner crystal can form if the electron-phonon interaction parameter α is larger than a critical value α_c and the electron density is lower than a critical value n_c.[10] The oscillations of the polarons in their self-trapping potential can generate a dipole-dipole interaction that destroys the crystal. The phonons undergo an instability due to the effect of the Lorentz local field, observable through signatures on the optical conductivity.[10] Moreover, in the dilute large polaron system, the softening of the phonon mode is the source of instability *versus* the formation of polaron pairs.[11] In the Hartree-Fock approximation and in the polaron Pekar model[12] the formation of strings[13] has been studied. Finally, the cluster formation in a dilute polaron gas has recently been shown[14] using the Vlasov nonlocal statistical approach[15] and a first preliminary result on the periodic phase in metal-ammonia systems has been given.[16]

In this work we study the instability of a dilute polaron gas toward the formation of a periodic phase depending on temperature, polaron density, dielectric parameters of the polar medium and strength of the electron-phonon coupling. This is done using the Bogolyubov method of the equilibrium distribution functions.[17] In this method a sequence of functions F_s for $s=1,2,\ldots$, is introduced; each function F_s depends on s position vectors $\boldsymbol{r}_1,\boldsymbol{r}_2,\ldots,\boldsymbol{r}_s$ and it determines the probability density of finding s particles in the unit volume of the configuration space of s particles. The knowledge of the distribution functions enables to define the spatial distribution of the system and, in particular, to find the thermodynamic instability conditions of an homogeneous and isotropic system of polarons toward a structured state. We are going to show that, depending on the polaron density, the electron-phonon coupling and the polarity parameters, the existence and stability of the periodic phase of polarons can only occur in a finite range of temperature. We shall see that the lower-temperature bound is determined by the kinetics of the system, while the upper bound by its thermodynamical equilibrium. Moreover, critical values of the electron-phonon

TABLE I. The parameters of the pair interpolaron potential [Eq. (3)]. $a_0^*=\hbar^2\varepsilon^*/e^2m^*$ is the effective Bohr radius.

$\varepsilon^*/\varepsilon_\infty$	a	$b\cdot a_0^{*2}$	$c\cdot a_0^{*2}$	d	$\gamma\cdot a_0^*$	$\delta\cdot a_0^*$	ρ_0/a_0^*
1.00	-7.920×10^{-4}	6.850×10^{-3}	-6.770×10^{-3}	-7.750×10^{-3}	0.300	0.150	7.500×10^{-3}
1.02	-6.000×10^{-4}	6.838×10^{-3}	-6.790×10^{-3}	-4.097×10^{-3}	0.284	0.160	1.525×10^{-2}
1.05	-1.907×10^{-5}	6.829×10^{-3}	-6.798×10^{-3}	-4.400×10^{-3}	0.254	0.194	2.250×10^{-2}
1.08	-5.175×10^{-5}	6.793×10^{-3}	-6.814×10^{-3}	1.000×10^{-3}	0.250	0.202	2.550×10^{-2}
1.10	-7.950×10^{-7}	6.675×10^{-3}	-6.815×10^{-3}	1.400×10^{-3}	0.250	0.232	2.750×10^{-2}

coupling and of the polarity parameters exist: under the critical conditions the temperature range reduces to zero and a stable, periodic phase cannot form.

The paper is organized as follows. In Sec. II we introduce the model and discuss the basic equations which allow the calculation of the equilibrium distribution functions. Next, the condition of existence of the periodic phase and its features are presented and discussed. In Sec. III we consider the stability in time of the polaron gas, discussed within the Bogolyoubov-Vlasov kinetic equation, which establishes the time evolution of the distribution function. In Sec. IV we compare the total energies of the periodic and homogeneous phases, giving a final indication that the periodic phase does actually appear provided that the conditions of existence and stability are met. Finally, in Sec. V we draw some conclusions.

II. THE PERIODIC PHASE

A. The model

We suppose to have a dilute polaron system in mechanical and thermodynamical equilibrium for which the interaction between two polarons $\Phi(\boldsymbol{r})$, where $\boldsymbol{r}$ is the relative distance of the two particles, is supposed to be known. The spatial configuration of the system can be completely characterized if the functions $F_s(\boldsymbol{r}_1,\boldsymbol{r}_2,\ldots,\boldsymbol{r}_s)$, with $s=1,2,3,\ldots$, are known. F_s is the probability density to find the particles in the unit volume of the configuration space with s dimensions. These functions are symmetric with respect to permutation of the variables. Taking into account the forces acting on the particles due to the internal and external fields, the distribution functions F_s satisfy, in the thermodynamic limit ($N\to\infty$, $V\to\infty$ and $N/V=n$ finite), the equations

$$\vartheta\frac{\partial F_s}{\partial r_1^q}+\frac{\partial[U_s+\psi(\boldsymbol{r}_1)]}{\partial r_1^q}F_s+n\int\frac{\partial\Phi(|\boldsymbol{r}_1-\boldsymbol{r}_{s+1}|)}{\partial r_1^q}F_{s+1}d\boldsymbol{r}_{s+1}=0,\quad q=x,y,z, \tag{1}$$

where $\vartheta=k_BT$, $U_s=\Sigma_{1\leqslant i<j\leqslant s}\Phi(|\boldsymbol{r}_i-\boldsymbol{r}_j|)$ is the potential energy of system and $\psi(\boldsymbol{r})$ the energy due to the external electrostatic field [see Eq. (6a)]. The distribution functions satisfy the relation

$$\lim_{V\to\infty}\frac{1}{V}\int F_{s+1}(\boldsymbol{r}_1,\boldsymbol{r}_2,\ldots,\boldsymbol{r}_{s+1})d\boldsymbol{r}_{s+1}=F_s(\boldsymbol{r}_1,\boldsymbol{r}_2,\ldots,\boldsymbol{r}_s),\quad s\geqslant1. \tag{2}$$

In all the calculations, we will use the potential Φ obtained in the strong coupling bipolaron Pekar theory adapted to give reliable results also in the intermediate coupling regime ($\alpha>5$, where α is the electron-phonon coupling constant in the large polaron theory).[18,19] This potential is coherent with that obtained within other approaches, for instance, the Lee-Low-Pines[20] and path-integral techniques.[21] As it is known, the interpolaron potential is long ranged and repulsive at large distances between polarons, attractive at intermediate distances (i.e., at distance near the polaron radius) and again repulsive at short distance.[18,19] The potential $\Phi(r)$ can be approximated by the following analytical form (see, for instance, Ref. 19):

$$\frac{\Phi(r)}{2\alpha^2\hbar\omega_0}=\{d+cr^2[a+b(r-\rho_0)^2][1-\exp(-\gamma r)]\}\exp(-\delta r)+\left(\frac{\varepsilon^*}{\varepsilon_\infty}-1\right)[1-\exp(-\delta r)]\frac{a_0^*}{r}, \tag{3}$$

where $a_0^*=\hbar^2\varepsilon^*/e^2m^*$ is the effective Bohr radius and the coefficients a, b, c, d, ρ_0, γ, and δ depend on $\varepsilon^*/\varepsilon_\infty$, where $\varepsilon^{*-1}=\varepsilon_\infty^{-1}-\varepsilon_s^{-1}$ and ε_s and ε_∞ are the static and high frequency dielectric constants. Φ depends also on the electron-phonon coupling constant $\alpha=(1/2\varepsilon^*)(e^2/\hbar\omega_0)(2m^*\omega_0/\hbar)^{1/2}$, where ω_0 is the optical longitudinal phonon frequency. Moreover, the electron and the phonon quantum exchange and the electron correlations are included with the virial theorem imposed as an additional condition. In Table I the above coefficients are reported for five values of $\varepsilon^*/\varepsilon_\infty$. Since $\varepsilon^*/\varepsilon_\infty=(1-\varepsilon_\infty/\varepsilon_s)^{-1}$, the medium polarity is a decreasing function of $\varepsilon^*/\varepsilon_\infty$. For instance, $\varepsilon^*/\varepsilon_\infty=1$ corresponds to a strongly polar medium ($\varepsilon_\infty/\varepsilon_s=0$), while for $\varepsilon^*/\varepsilon_\infty=1.10$ one has $\varepsilon_\infty/\varepsilon_s=0.091$ (i.e., the high-frequency dielectric constant is ten times smaller that the static one). In Fig. 1(a) the function $\Phi(r)$ is shown. It is seen that the region in which $\Phi(r)<0$ becomes narrower on increasing $\varepsilon_\infty/\varepsilon_s$, in agreement with the previous remarks. Moreover, $\Phi(r)\to0$ as $r\to\infty$. This means that the zero of the energy is that of two free polarons in the strong coupling limit, i.e., $E_p=-(2\alpha^2/3\pi)\hbar\omega_0$. In the calculation the free-electron mass is taken as the electron effective mass and we take $\hbar\omega_0=0.30$ eV. Finally, the system electrical neutrality is ensured by a positive charge density $\rho_0=-en$ ($e<0$ is the electron charge).

The single- and two-particle distribution functions $F_1(\boldsymbol{r}_1)$ and $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ satisfy the two equations

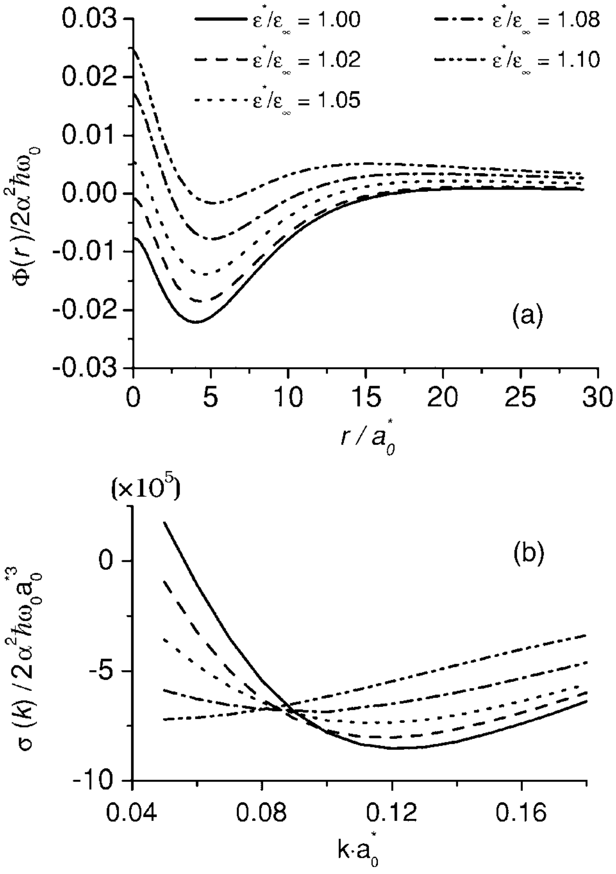

FIG. 1. (a) The interpolaron pair potential as a function of the relative distance and (b) the Fourier transform $\sigma(k)$ of the collective potential $K(r)$ shown for different values of $\varepsilon^*/\varepsilon_\infty$ (we fix $\alpha=15$ and $n=10^{18}$ cm^{-3}).

$$\vartheta\frac{\partial F_1(\boldsymbol{r}_1)}{\partial r_1^q}+F_1(\boldsymbol{r}_1)\frac{\partial\psi(\boldsymbol{r}_1)}{\partial r_1^q}+n\int\frac{\partial\Phi(|\boldsymbol{r}_1-\boldsymbol{r}_2|)}{\partial r_1^q}F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)d\boldsymbol{r}_2=0 \tag{4}$$

and

$$\vartheta\frac{\partial F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)}{\partial r_1^q}+\frac{\partial[\Phi(|\boldsymbol{r}_1-\boldsymbol{r}_2|)+\psi(\boldsymbol{r}_1)]}{\partial r_1^q}F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)+n\int\frac{\partial\Phi(|\boldsymbol{r}_1-\boldsymbol{r}_3|)}{\partial r_1^q}F_3(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_3)d\boldsymbol{r}_3=0, \tag{5}$$

whereas the electrostatic energy $\psi(\boldsymbol{r})$ is given by

$$\nabla^2\psi(\boldsymbol{r})=-\frac{4\pi e}{\varepsilon_s}[\rho_1(\boldsymbol{r})+\rho_0], \tag{6a}$$

where $\rho_1(\boldsymbol{r})=enF_1(\boldsymbol{r})$ and $\rho_0=-en$ are the densities of the nonhomogeneous polarons distribution and of the homogeneous positive background, respectively. The set of equations (4), (5), and (6a) is not complete because the function $F_3(\boldsymbol{r}_1,\boldsymbol{r}_2,\boldsymbol{r}_3)$ is unknown. In the following the contribution of the integral containing F_3 (i.e., three-particles correlation effects) will be neglected. The distribution functions $F_1(\boldsymbol{r}_1)$ and $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ obey the normalization conditions

$$\lim_{V\to\infty}\frac{1}{V}\int F_1(\boldsymbol{r})d\boldsymbol{r}=1,\quad \lim_{V\to\infty}\frac{1}{V^2}\int F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)d\boldsymbol{r}_1d\boldsymbol{r}_2=1. \tag{6b}$$

Equation (4) can be solved for the one-particle distribution function $F_1(\boldsymbol{r})$ if the two-particle distribution function $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ is known. The knowledge of $F_1(\boldsymbol{r}_1)$ and $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ allows us to define an equilibrium state of a statistical system made of interacting particles. As a first step, we decouple Eqs. (4) and (5) by assuming

$$F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)=F_1(\boldsymbol{r}_1)F_1(\boldsymbol{r}_2). \tag{7}$$

This ansatz is well verified in the very dilute system[17] when the relative distance between the particles is larger than the polaron radius.[12] Although Eq. (7) completely neglects the correlation between the particles, it has the advantage of allowing a self-consistent solution of Eqs. (4) and (5).[15,22] Obviously, the increase of the polaron density makes the approximation of Eq. (7) incorrect. Therefore, as a second step, we take into account the particle-particle correlation writing [let us note that $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ is invariant with respect to the permutation of $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$]

$$F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)=G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)F_1(\boldsymbol{r}_1)F_1(\boldsymbol{r}_2). \tag{8}$$

Here $G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)$ is the radial pair correlation function, satisfying the boundary condition $G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)\to 1$ as $|\boldsymbol{r}_1-\boldsymbol{r}_2|\to\infty$. It can be calculated using Eqs. (8) and (5), with $F_1(r)=\text{const}$ (i.e., we calculate the correlation function of the homogeneous polaron gas, for which the electrostatic energy ψ is zero). Thus, we are left with the following equation for $G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)$,

$$\vartheta\frac{\partial G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)}{\partial r_1^q}+\frac{\partial\Phi(|\boldsymbol{r}_1-\boldsymbol{r}_2|)}{\partial r_1^q}G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)=0. \tag{9}$$

The solution is

$$G_2(r)=C\exp[-\Phi(r)/\vartheta], \tag{10}$$

where, taking into account the boundary condition, one has $C=1$.

With the previous ansatz the effect of the density n on the distribution function $F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)$ is ruled out. This can be included if, as a third and last step, we assume

$$F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)=F_1(\boldsymbol{r}_1)F_1(\boldsymbol{r}_2)\{1+\varepsilon_0[G_2(|\boldsymbol{r}_1-\boldsymbol{r}_2|)-1]\}, \tag{11}$$

where $\varepsilon_0=r_0^3/v$ is a dilution parameter, r_0 is the effective polaron size,[11] and $v=V/N$ the average volume per particle. Equation (11) for F_2 verifies the following conditions: (i) it reduces to Eq. (7) if the relative distance between the particles becomes large; (ii) it reduces again to Eq. (7) if the system is very dilute ($\varepsilon_0 \ll 1$); (iii) it takes into account the increase of the correlation on increasing n. Combining Eqs. (11) and (4) and taking into account the normalization condition (6b) for $F_1(\boldsymbol{r})$, the nonlinear integral equation

$$\ln[F_1(\boldsymbol{r}_1)\lambda] + \frac{\psi(\boldsymbol{r}_1)}{\vartheta} + \frac{n}{\vartheta}\int K(|\boldsymbol{r}_1-\boldsymbol{r}_2|)F_1(\boldsymbol{r}_2)d\boldsymbol{r}_2 = 0 \tag{12}$$

is found. Here $K(r)=(1+\varepsilon_0)^{-1}(\Phi(r)+(\vartheta\varepsilon_0/n_0)\{1-\exp[-\Phi(r)/\vartheta]-\Phi(r)/\vartheta\})$ is the effective interaction potential,

$$n_0 = \left[\frac{4\pi}{V}\int_0^\infty \{\exp[-\Phi(r)/\vartheta]-1\}r^2dr\right],$$

and λ is a Lagrange multiplier introduced to take into account the normalization condition (6b). Since $K(r)=\Phi(r)$ if $\varepsilon_0=0$, $K(r)$ can be viewed as the interpolaron potential with the inclusion of correlation effects. We note that the correction to $\Phi(r)$ included in $K(r)$ has not a definite sign. This is because $1-\exp[-\Phi(r)/\vartheta]-\Phi(r)/\vartheta$ is always negative, whereas the sign of n_0 depends on the temperature. We conclude that the correlation can either enhance or reduce the polaron potential $\Phi(r)$. Such collective effects have been shown, within many-polaron theories at $T=0$, to influence the bipolaron binding energy in both the two- and three-dimensional case[23,24] as well as the optical absorption.[25] It is found that the bipolaron self-energy is enhanced for electron densities up to about 10^{12} cm^{-2} in the two-dimensional case and $10^{18}-10^{19}$ cm^{-3} in the three-dimensional one. For higher density the self-energy goes to zero, indicating that the collective effects destroy the bipolaron formation. The optical absorption, instead, shows a monotonic variation with the density.

The function $F_1(r)=1$, which satisfies the normalization condition (6a) and corresponds to a uniform spatial distribution of polarons, is a solution of Eq. (12) if $\lambda=\lambda_0$ with λ_0 solution of $\ln(\lambda_0)+(4\pi n/\vartheta)\int K(r)r^2dr=0$. Equation (12) is a nonlinear integral equation of Hammerstein type that can present the phenomenon of bifurcation, i.e., the possibility to have simultaneously two different independent solutions.[26] We are going to show that Eq. (12), under suitable conditions on temperature, density of polarons, electron-phonon coupling constant and dielectric properties, has solutions different from the trivial one. We consider explicitly only the case of the temperature to see that there exists a critical temperature $T_{\text{cr}}^{(1)}=\vartheta_0/k_B$ near which a solution different from $F_1=1$ exists. In other words, the critical temperature $T_{\text{cr}}^{(1)}$ corresponds to bifurcation points of the solutions of the integral equation (12). Using the method of the successive approximations,[26] we expand F_1, λ, ψ, and ϑ as function of the quantity $(\vartheta_0-\vartheta)^{1/2}/\vartheta_0^{1/2}$ obtaining

$$F_1(\boldsymbol{r}) = \chi_0 + \frac{(\vartheta_0-\vartheta)^{1/2}}{\vartheta_0^{1/2}}\chi_1(\boldsymbol{r}) + \frac{(\vartheta_0-\vartheta)}{\vartheta_0}\chi_2(\boldsymbol{r}) + \cdots,$$

$$\lambda = \lambda_0 + \frac{(\vartheta_0-\vartheta)^{1/2}}{\vartheta_0^{1/2}}\lambda_1 + \frac{(\vartheta_0-\vartheta)}{\vartheta_0}\lambda_2 + \cdots,$$

$$\frac{1}{\vartheta} = \frac{1}{\vartheta_0} + \frac{(\vartheta_0-\vartheta)}{\vartheta_0^2} + \cdots,$$

$$\psi(\boldsymbol{r}) = \psi_0 + \frac{(\vartheta_0-\vartheta)^{1/2}}{\vartheta_0^{1/2}}\psi_1(\boldsymbol{r}) + \frac{(\vartheta_0-\vartheta)}{\vartheta_0}\psi_2(\boldsymbol{r}) + \cdots,$$

$$\psi_0 = 0. \tag{13}$$

Using the expansion (13) in Eq. (12) and collecting terms with equal powers of $(\vartheta_0-\vartheta)^{1/2}/\vartheta_0^{1/2}$, we obtain to the zero order

$$\ln\lambda_0 + \frac{n}{\vartheta_0}\int K(|\boldsymbol{r}_1-\boldsymbol{r}_2|)d\boldsymbol{r}_1 = 0, \quad \lim_{V\to\infty}\frac{1}{V}\int \chi_0 d\boldsymbol{r} = 1 \tag{14}$$

and, to the first order,

$$\frac{\lambda_1}{\lambda_0} + \chi_1(\boldsymbol{r}_1) = -\frac{n}{\vartheta_0}\int K(|\boldsymbol{r}_1-\boldsymbol{r}_2|)\chi_1(\boldsymbol{r}_2)d\boldsymbol{r}_2 - \frac{\psi_0\chi_1(\boldsymbol{r}_1)}{\vartheta_0} - \frac{\psi_1(\boldsymbol{r}_1)\chi_0}{\vartheta_0},$$

$$\lim_{V\to\infty}\frac{1}{V}\int \chi_1(\boldsymbol{r})d\boldsymbol{r} = 0. \tag{15}$$

Equations (14) are satisfied if $\chi_0=1$ with the previously calculated value of λ_0. In Eqs. (15) we have $\lambda_1=0$ because $\lim_{V\to\infty}(1/V)\int\chi_1(r)r^2dr=0$ and $\psi_0=0$. This means that the highest critical bifurcation temperature ϑ_0 comes from the lowest eigenvalue $1/\vartheta_0$ of the linear Fredholm integral equation

$$\chi_1(\boldsymbol{r}_1) + \frac{\psi_1(\boldsymbol{r}_1)}{\vartheta_0} = -\frac{n}{\vartheta_0}\int K(|\boldsymbol{r}_1-\boldsymbol{r}_2|)\chi_1(\boldsymbol{r}_2)d\boldsymbol{r}_2. \tag{16}$$

Since the spatial Fourier transform of $\psi_1(\boldsymbol{r})$ and of $\chi_1(\boldsymbol{r})$ are proportional, the Fourier transformed integral equation (16) is an homogeneous linear integral equation with eigenvalue $1/\vartheta_0$. We remark that the bifurcation phenomenon can concern many new physical structures, as the formation of clusters or periodic phases. Since ϑ_0 is a function of the parameters α, $\varepsilon^\star/\varepsilon_\infty$, and n, which enter in the equation, it could occur that, depending on the bifurcation temperature, different structured states are obtained, all arising from the uniformly distributed polaron gas. In this work we consider only the possibility of having periodic solution of Eq. (16) of the type $\chi_1(\boldsymbol{r})\approx\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$. By substitution of this solution in Eq. (16), we find that the condition which assures the existence of eigenvalues is

$$\mu = -\frac{n\sigma(k)}{\vartheta_0\left[1+\dfrac{4\pi e^2 n}{\varepsilon_s(k^2+\kappa^2)\vartheta_0}\right]} \geqslant 1, \tag{17}$$

where $\sigma(k)=\int K(r)\exp(i\boldsymbol{k}\cdot\boldsymbol{r})d\boldsymbol{r}$ is the form factor and $\kappa=(4\pi e^2 n/\varepsilon_s k_B T)^{1/2}$ is the reciprocal Debye screening length for long-range Coulomb interactions. The condition $\mu=1$ gives the bifurcation point of the integral equation where the polaron periodic phase starts. To obtain the largest critical temperature $T_{\text{cr}}^{(1)}$ we must find, in the region where $\sigma(k)<0$, the value k_{opt} such that $\sigma(k)$ assumes the minimum value.

For this value of k, the highest critical temperature $T_{\mathrm{cr}}^{(1)}$ is found considering the value of $1/\vartheta_0$ which satisfies the condition $\mu=1$. As it is shown in Fig. 1(b), for a fixed value of α, $\sigma(\kappa)$ can be negative. For a medium that has a strong polarity k_{opt} is finite, while in the opposite limit it decreases, i.e., the periodic phase tends to disappear. This value of the wave vector corresponds to the spatial period $d=2\pi/k_{\mathrm{opt}}$ of the polaron distribution. We stress that we have not found a specific crystal structure for the polaron structured state, but only a distance d between planes on which the function $F_1(\boldsymbol{r})$ takes the same value. We do not calculate the effective crystal structure of the structured phase. This is a more difficult task that could be done, in principle, in the framework of the present theory, but this calculation is beyond the aim of this work. We stress only that cubic structures should be a possible lattice as a consequence of the dependence of the potential $K(r)$ on the modulus of the polarons relative distance.

B. Results

Equation (17) allows us to establish the appearance of the spatially periodic component in the uniform polarons distribution. $\sigma(k)$ depends on n, on the electron-phonon coupling constant α, on the dielectric parameter $\varepsilon^*/\varepsilon_\infty$ of the polar medium and on the temperature T. As soon as T becomes less than $T_{\mathrm{cr}}^{(1)}=\vartheta_0/k_B$, a space periodic component in the polarons distribution spontaneously appears. If the electron-phonon coupling constant α decreases, ϑ_0 is reduced; this decrease can be obtained, with fixed ω_0, either taking fixed $\varepsilon^*/\varepsilon_\infty$ and increasing ε_∞ or taking fixed ε_∞ and increasing $\varepsilon^*/\varepsilon_\infty$. In both the cases, the region in which $\Phi(r)$ is negative becomes narrower [Fig. 1(a)] and k_{opt} tends to zero [Fig. 1(b)] giving an indication that the periodic phase appears with major difficulty.

A simple way to study the variation of the bifurcation temperature is taking fixed α, ω_0, and the polarons density n, but changing $\varepsilon^*/\varepsilon_\infty$. We find that the critical temperature $T_{\mathrm{cr}}^{(1)}$ is a decreasing function of $\varepsilon^*/\varepsilon_\infty$, as shown in Fig. 2(a), where $n=10^{18}$ cm^{-3}, $\hbar\omega_0=0.30$ eV, and different values of α are considered. We note that (a) each curve with fixed α is a decreasing function of $\varepsilon^*/\varepsilon_\infty$ in agreement with the fact that the increase of $\varepsilon^*/\varepsilon_\infty$ means that the medium becomes less polar (b) for a fixed value of $\varepsilon^*/\varepsilon_\infty$, $T_{\mathrm{cr}}^{(1)}$ is an increasing function of α in agreement with the above remark.

In Fig. 3 we show the dependence of k_{opt} on $\varepsilon^*/\varepsilon_\infty$ for different values of α, with $n=10^{18}$ cm^{-3} and $T=T_{\mathrm{cr}}^{(1)}$. We find that, for any fixed α, k_{opt} is a slowly decreasing function of $\varepsilon^*/\varepsilon_\infty$ when the medium is strongly polar and has a value independent of α as $\varepsilon^*/\varepsilon_\infty \to 1$. Furthermore, for each curve a specific value $(\varepsilon^*/\varepsilon_\infty)_1$ of $\varepsilon^*/\varepsilon_\infty$ is found, such that, for $\varepsilon^*/\varepsilon_\infty > (\varepsilon^*/\varepsilon_\infty)_1$, k_{opt} quickly tends to zero. We see also that $(\varepsilon^*/\varepsilon_\infty)_1$ strongly depends on α. From the physical point of view, we see that if the medium becomes less polar, the periodic phase disappears and the system becomes homogeneous because the spatial periodicity tends to infinity.

In Fig. 2(b) we show, at a fixed polaron density $n=10^{18}$ cm^{-3}, the dependence of the critical temperature $T_{\mathrm{cr}}^{(1)}$ on α, for different values of $\varepsilon^*/\varepsilon_\infty$. Each of these curves defines the separation line between the two phases (with respect to the temperature): in the upper region the polaron system is homogeneous while in the lower one it shows a periodic phase. An important point which can be inferred from the figure is that the periodic phase cannot appear for any α. Each curve starts at a specific value of α, that indicates the lowest value of α, α_{cr}, compatible with the struc-

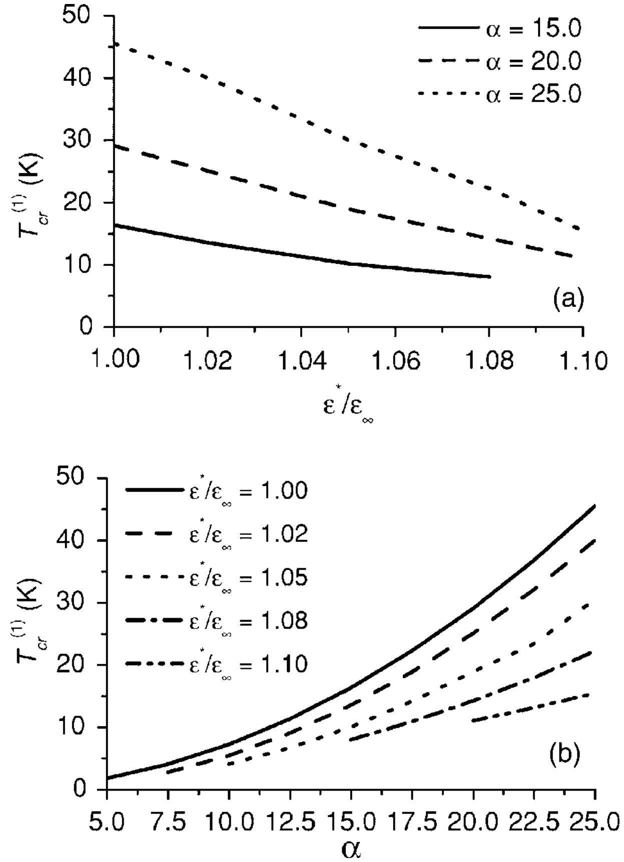

FIG. 2. The dependence of the critical temperature $T_{\mathrm{cr}}^{(1)}$ on (a) $\varepsilon^*/\varepsilon_\infty$ for different values of the electron-phonon coupling constant α and (b) the electron-phonon coupling constant α for different values of $\varepsilon^*/\varepsilon_\infty$ (we fix $n=10^{18}$ cm^{-3}).

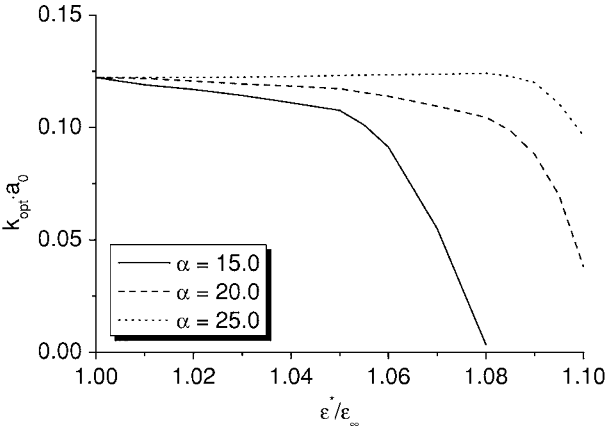

FIG. 3. The dependence of the wave vector k_{opt} on $\varepsilon^*/\varepsilon_\infty$ for different values of the electron-phonon coupling constant α (we fix $n=10^{18}$ cm^{-3} and $T=T_{\mathrm{cr}}^{(1)}$).

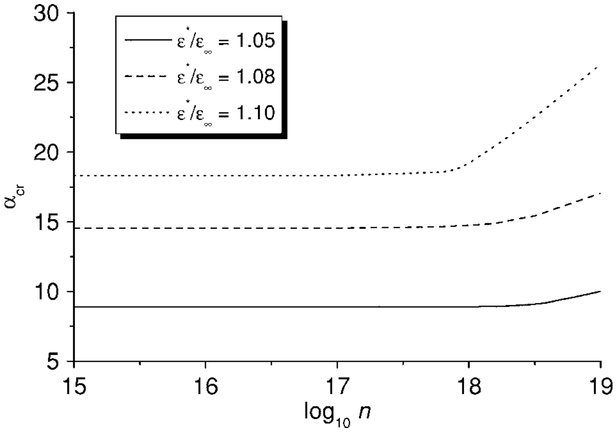

FIG. 4. The dependence of the critical value of the electron-phonon coupling constant α_{cr} on the density n for different values of $\varepsilon^*/\varepsilon_\infty$.

tured phase. For example, if $\varepsilon^*/\varepsilon_\infty=1.10$, the line starts at $\alpha=20$. This means that the periodic phase appears only for $\alpha \geqslant \alpha_{\mathrm{cr}}=20$; at the same polaron density and if $\varepsilon^*/\varepsilon_\infty=1.05$, the periodic phase appears for $\alpha \geqslant 8.9$ and $\alpha_{\mathrm{cr}}=8.9$. This strong decrease of α_{cr} is a consequence of the large increase of the ionic polarity of the medium.

In Fig. 4 we show α_{cr} as function of the polaron density n for three values of $\varepsilon^*/\varepsilon_\infty$. We see that up to $n \approx 10^{18}$ cm^{-3}, α_{cr} is nearly constant for each fixed value $\varepsilon^*/\varepsilon_\infty$; for larger density, α_{cr} increases more quickly and the increasing rate is larger for less polar media. We note that the collective effects begin to be effective for densities of about 10^{18} cm^{-3}, as it was found in Ref. 4.

Finally, in Fig. 5 we show the critical temperature $T_{\mathrm{cr}}^{(1)}$ as function of n for a fixed value of $\varepsilon^*/\varepsilon_\infty$ and for several values of α. In the considered range of polaron density, we see that $T_{\mathrm{cr}}^{(1)}$ increases with n. Also in this case we find that the collective effects appear for density around 10^{18} cm^{-3}.

The results of this section would show that, at a given temperature, there is no limit to the formation of the periodic phase provided that $\alpha \geqslant \alpha_{\mathrm{cr}}$. Nevertheless, as we are going to show in the next section, a further condition arises by requiring the stability in time of the periodic phase.

III. INSTABILITY OF THE HOMOGENEOUS POLARON SYSTEM

In this section we discuss the stability in the time of the periodic phase. This can be done in the framework of the Bogolyubov-Vlasov kinetic equation[15] which, for central forces, is

$$\frac{\partial f(\boldsymbol{r},\boldsymbol{v},t)}{\partial t} + \boldsymbol{v} \cdot \boldsymbol{\nabla}_r f(\boldsymbol{r},\boldsymbol{v},t) = \frac{1}{M} \boldsymbol{\nabla}_v f(\boldsymbol{r},\boldsymbol{v},t) \cdot \boldsymbol{\nabla}_r \left[\int\!\!\int_{(\infty)} \Phi(|\boldsymbol{r}-\boldsymbol{r}'|) f(\boldsymbol{r}',\boldsymbol{v}',t)\, d\boldsymbol{r}'\, d\boldsymbol{v}' + \psi(\boldsymbol{r}) \right], \tag{18}$$

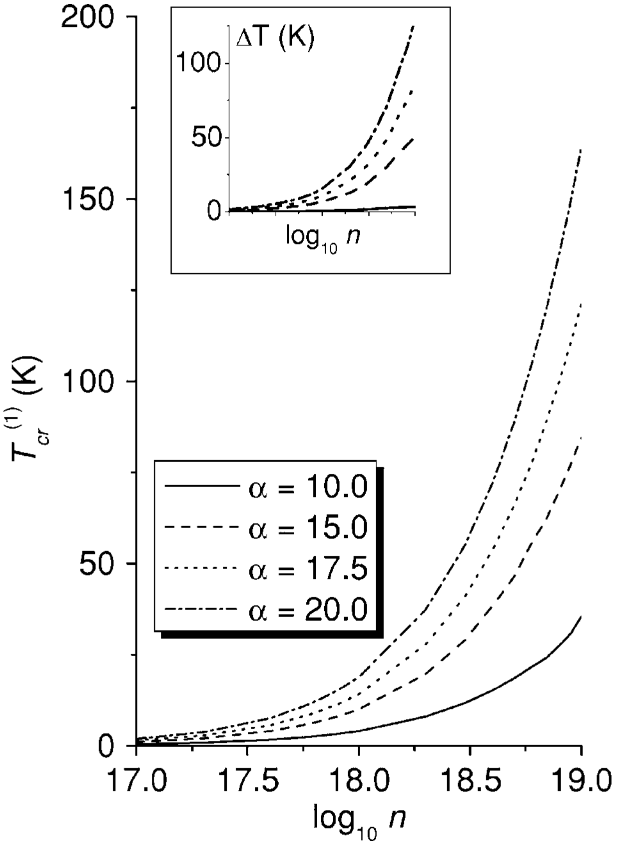

FIG. 5. The dependence of the critical temperature $T_{\mathrm{cr}}^{(1)}$ on the density n for different values of the electron-phonon coupling constant α (we fix $\varepsilon^*/\varepsilon_\infty=1.05$). The inset shows the dependence on the polaron density of the temperature interval $\Delta T=T_{\mathrm{cr}}^{(1)}-T_{\mathrm{cr}}^{(2)}$ (see Sec. III) for the same values of α.

where $f(\boldsymbol{r},\boldsymbol{v},t)$ is the polaron kinetic distribution function, $M=0.0229\alpha^4 m^*$ the polaron mass and $\boldsymbol{v}$ its velocity. Equation (18) describes the changes in time of the distribution function in the single-particle phase space. In the stationary case the kinetic distribution function is given by the Boltzmann function $f_0(\boldsymbol{v}^2)=n(M/2\pi\vartheta)^{3/2}\exp(-M\boldsymbol{v}^2/2\vartheta)$. We are going to show that introducing any small perturbation $\varphi(\boldsymbol{r},\boldsymbol{v},t)$ to the distribution function, under suitable conditions, the solution of Eq. (18) results in an instability in time. Near the bifurcation point, we look for a solution of Eq. (18) in the form

$$f(\boldsymbol{r},\boldsymbol{v},t) = f_0(\boldsymbol{v}^2) + \varphi(\boldsymbol{r},\boldsymbol{v},t), \quad |\varphi| \ll f_0. \tag{19}$$

We assume that $\varphi(\boldsymbol{r},\boldsymbol{v},t)$ is a small perturbation of the stationary homogeneous particle distribution $f_0(\boldsymbol{v}^2)$, so that we can retain only the linear terms in Eq. (18). We obtain

$$\frac{\partial \varphi(\boldsymbol{r},\boldsymbol{v},t)}{\partial t} + \boldsymbol{v} \cdot \boldsymbol{\nabla}_r \varphi(\boldsymbol{r},\boldsymbol{v},t) = \frac{1}{M} \boldsymbol{\nabla}_v f_0(\boldsymbol{v}^2) \cdot \boldsymbol{\nabla}_r \left[\int\!\!\int_{(\infty)} \Phi(|\boldsymbol{r}-\boldsymbol{r}'|) \varphi(\boldsymbol{r}',\boldsymbol{v}',t)\, d\boldsymbol{r}'\, d\boldsymbol{v}' + \psi \right]. \tag{20}$$

Furthermore, $\psi=0$ for small deviations from the homogeneous distribution of particles. The solution of Eq. (20) provides the spatial distribution of the particles $\rho(\boldsymbol{r},t)=\int_{(\infty)}\varphi(\boldsymbol{r},\boldsymbol{v},t)d\boldsymbol{v}$. We look for a solution $\rho(\boldsymbol{r},t)$ in the form $\rho(\boldsymbol{r},t)=\Sigma_k\rho_k(t)\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$. By assuming the initial condition $\varphi(\boldsymbol{r},\boldsymbol{v},t_0)=\Sigma_k\varphi_k(\boldsymbol{v},t_0)\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$, with $\varphi_k(\boldsymbol{v},t_0)=a_kf_0(\boldsymbol{v}^2)$, we obtain (see Appendix A)

$$\rho_k(t)=a_kn\exp(-kt/\beta)\mathrm{ch}(\Omega t),\tag{21}$$

where $\Omega^2=-\sigma_0(k)nk^2/M$ is the square of a frequency, $\sigma_0(k)$ is the Fourier transform of the interpolaron potential (see Appendix A) and $\beta=[M/2\vartheta]^{1/2}$. If it occurs that

$$-\frac{\sigma_0(k)n}{\vartheta}>1,\tag{22}$$

then Ω is real and $\rho_k(t)$ increases with time clearly indicating an instability of the periodic polaron system.

We stress that this result has to be combined with that in the previous section to assess the existence and stability of the periodic phase. Indeed, Eq. (17) gives the condition for the existence of a periodic phase while Eq. (22) determines the instability in time of the Fourier component $\rho_k(t)$ of the polaronic distribution function. This means that if Eq. (22) is satisfied for $k=k_{\mathrm{opt}}$, Eq. (17) would foresee the existence of the periodic phase, but this last one is not stable.

The previous result gives an indication for the possibility of nonuniform oscillations in the homogeneous gas. This suggests that the formation of the periodic phase corresponds to a wavelike solution of Eq. (20) (i.e., such that the nonuniform oscillations are periodic), which can be sustained with no damping. Therefore, we shall now look for a solution of Eq. (20) in the form $\varphi(\boldsymbol{r},\boldsymbol{v},t)=\varphi_k(\boldsymbol{v})\exp[i\boldsymbol{k}\cdot\boldsymbol{r}+(i\Omega+\gamma)t]$, where it is assumed that the frequency of the propagating wave is the previously defined Ω. A damping coefficient γ has been introduced. We shall see that $\varphi(\boldsymbol{r},\boldsymbol{v},t)$ does satisfy Eq. (20), but both real and purely imaginary values of γ are possible. In the first case the propagating wave will be damped out and the periodic phase cannot be sustained, because unstable in time. Therefore, the ultimate goal is that of finding the condition under which a solution with a purely imaginary γ is possible. Such a condition will define the kinetic stability of the periodic phase. By inserting $\varphi(\boldsymbol{r},\boldsymbol{v},t)$ in Eq. (20) we obtain the following operational equation for $\varphi_k(v)$:

$$\left(-iL-i\boldsymbol{k}\cdot\boldsymbol{v}+\frac{i\sigma_0(k)}{M}\boldsymbol{k}\cdot\boldsymbol{\nabla}_{\boldsymbol{v}}f_0\int_{(\infty)}d\boldsymbol{v}\right)\varphi_k(\boldsymbol{v})=0,\tag{23}$$

where $L=\Omega+i\gamma$.

The solution L of Eq. (23) is found if we determine the poles of the operator $\{-iL-i\boldsymbol{k}\cdot\boldsymbol{v}+[i\sigma_0(k)/M]\boldsymbol{k}\cdot\boldsymbol{\nabla}_{\boldsymbol{v}}f_0\int_{(\infty)}d\boldsymbol{v}\}^{-1}$. As shown in Appendix B, this is equivalent to solve the equation

$$\frac{\vartheta}{n\sigma_0(k)}=-1+2Z\exp(-Z^2)\int_0^Z\exp(s^2)ds,\tag{24}$$

where $Z=(1/k)(\Omega+i\gamma)(M/2\vartheta)^{1/2}$. As stated above, as far as γ is real, it assumes the role of a damping coefficient. On the other hand, if a purely imaginary solution γ can be found, the periodic solution for the particle distribution function has no damping and, therefore, is stable. At fixed polaron density n and electron-phonon coupling constant α, a purely imaginary solution $\gamma\neq0$ exists [as it follows from Eq. (B2)] only if $T_{\mathrm{cr}}^{(2)}<T<T_{\mathrm{cr}}^{(1)}$, the last temperature being calculated from Eq. (17) (see the previous section). $T_{\mathrm{cr}}^{(2)}$ is the lowest temperature compatible with a solution with purely imaginary γ. In Fig. 6(a) we show, as a function of the temperature and for different values of $\varepsilon^*/\varepsilon_\infty$ (we fix $n=10^{18}$ cm^{-3} and $\alpha=20$), the solution for $|\gamma|$ when γ is purely imaginary. In this way we define the region of existence of the periodic polaron phase. For temperatures $T<T_{\mathrm{cr}}^{(2)}$ and $T>T_{\mathrm{cr}}^{(1)}$ the periodic polaron phase does not appear; both $T_{\mathrm{cr}}^{(2)}$ and $T_{\mathrm{cr}}^{(1)}$ indicate the starting of furcated solutions of Eq. (12). It is found that the finite temperature range in which the periodic phase exists increases when the medium becomes more polar. It is also evident from Fig. 6(a) that there is an upper limit to $\varepsilon^*/\varepsilon_\infty$, $(\varepsilon^*/\varepsilon_\infty)_{\mathrm{cr}}$, such that if $\varepsilon^*/\varepsilon_\infty>(\varepsilon^*/\varepsilon_\infty)_{\mathrm{cr}}$ the periodic phase cannot appear. For the case shown in Fig. 6(a), we find that $(\varepsilon^*/\varepsilon_\infty)_{\mathrm{cr}}=1.12$. On the other hand, keeping fixed n

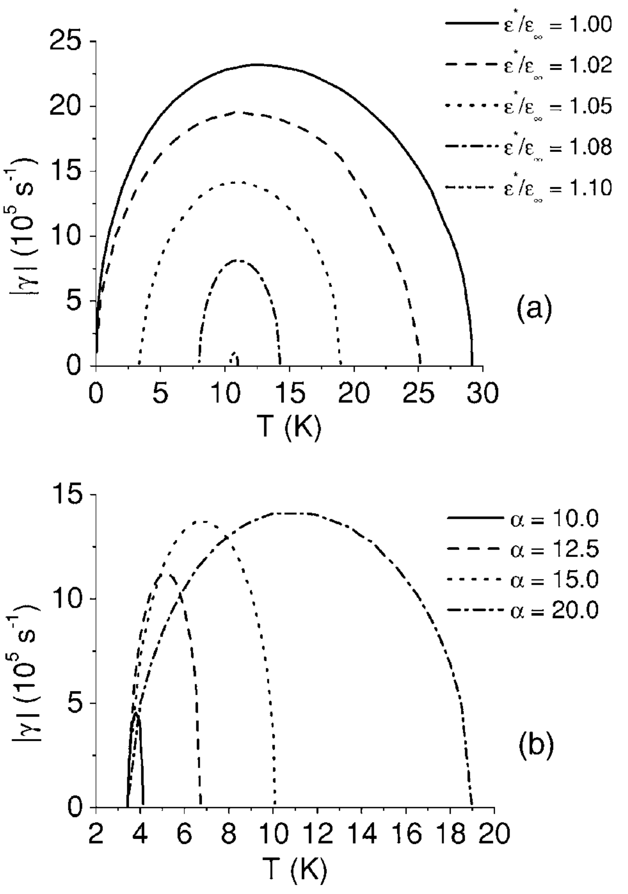

FIG. 6. The dependence of $|\gamma|(\gamma=i|\gamma|)$ on the temperature (a) for different values of $\varepsilon^*/\varepsilon_\infty$ (we fix $n=10^{18}$ cm^{-3} and $\alpha=20$) and (b) for different electron-phonon coupling constants α (we fix $n=10^{18}$ cm^{-3} and $\varepsilon^*/\varepsilon_\infty=1.05$). The two crossing points of each line with the horizontal axis define the temperature range in which the periodic phase can exist. For a given $\varepsilon^*/\varepsilon_\infty$ in (a) or α in (b) the periodic phase of the polaron distribution can occur only in the region delimited by the respective curve.

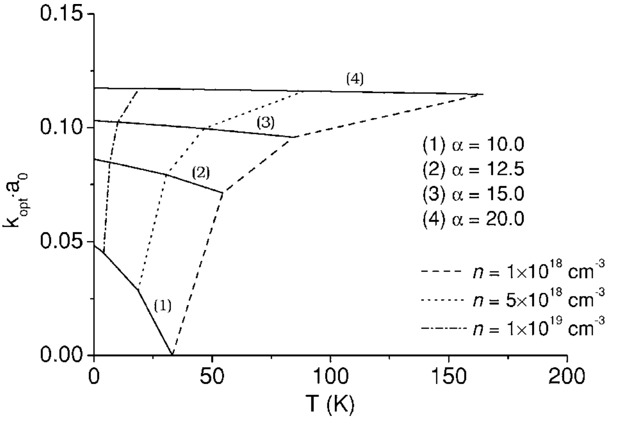

FIG. 7. The dependence of the wave number k_{opt} (solid lines) of the periodic phase on the temperature T and for different values of coupling constant α (we fix $\varepsilon^*/\varepsilon_\infty = 1.05$). The dashed, dot, and dashed-dot lines show the wave number k_{opt} at the critical temperature $T_{\text{cr}}^{(1)}$ for different values of n. In the region on the right of these lines the periodic polaron distribution cannot exist ($T > T_{\text{cr}}^{(1)}$).

$=10^{18}$ cm^{-3} and $\varepsilon^*/\varepsilon_\infty = 1.05$, we show in Fig. 6(b), the range of existence of the periodic phase for different values of α. In this case we find that $T_{\text{cr}}^{(2)}$ is the same for all the values of α, but $T_{\text{cr}}^{(1)}$ increases on increasing α, as it is was already shown in Fig. 2(b). Furthermore, Fig. 6(b) indicates that the temperature range in which the periodic phase exists decreases on decreasing α, making reliable the value of $\alpha_{\text{cr}} = 8.9$ shown in Fig. 2(b) ($\varepsilon^*/\varepsilon_\infty = 1.05$).

The variation of the temperature interval $\Delta T = T_{\text{cr}}^{(1)} - T_{\text{cr}}^{(2)}$ in which the periodic phase exists with the polaron density is reported in the inset of Fig. 5 (we fix $\varepsilon^*/\varepsilon_\infty = 1.05$). In the range of polaron density considered, it is an increasing function of n and, for a fixed density, an increasing function of α.

Finally, in Fig. 7 we show the dependence of k_{opt} on the temperature T for $\varepsilon^*/\varepsilon_\infty = 1.05$ and for different values of α. Furthermore, in the same figure the wave number k_{opt} is shown at the critical temperature for three different densities. We remark that if the periodic phase gets less stable (on decreasing α in Fig. 7), the periodicity length of the spatial polaron distribution increases.

The analysis we have performed brings out that we can fix a number of physical parameters $T_{\text{cr}}^{(1)}$, $T_{\text{cr}}^{(2)}$, α_{cr}, $(\varepsilon^*/\varepsilon_\infty)_{\text{cr}}$ which completely define the regions of existence of the periodic polaron phase, i.e., $T_{\text{cr}}^{(2)} < T < T_{\text{cr}}^{(1)}$, $\alpha > \alpha_{\text{cr}}$, and $\varepsilon^*/\varepsilon_\infty < (\varepsilon^*/\varepsilon_\infty)_{\text{cr}}$.

IV. TOTAL ENERGIES

It is clear that if all the found conditions on the existence and stability of the periodic phase are met, the possibility of formation of this phase can be questionable, because it can still be a metastable state. This means that the full assessment of the formation of the periodic phase requires a last step, namely the calculation of the total energy of the homogeneous and periodic systems. The system of polarons makes a transition to a state with periodic structure if this state has smaller energy in comparison with the homogeneous distribution. The total energy per polaron can be computed through

$$E = \frac{3}{2}k_B T + \frac{N^2}{2V^2}\int\int \Phi(|\boldsymbol{r}_1 - \boldsymbol{r}_2|)F_2(\boldsymbol{r}_1,\boldsymbol{r}_2)d\boldsymbol{r}_1 d\boldsymbol{r}_2$$
$$= \frac{3}{2}k_B T + U. \qquad (25)$$

The results show that in all the area of existence of the periodic distribution of polarons, this last one is energetically more favorable than the homogeneous one. As an example, in Fig. 8 we show the ratio $|U_{\text{periodic}}|/|U_{\text{hom}}|$ ($U<0$) for different values of $\varepsilon^*/\varepsilon_\infty$ as a function of the electron-phonon coupling constant. It is seen that the periodic phase is the lowest energy configuration in all the cases. Finally, we have also found that the ratio is practically independent on the polaron density n, which allows us to assess the formation of the periodic phase in all the range of considered densities.

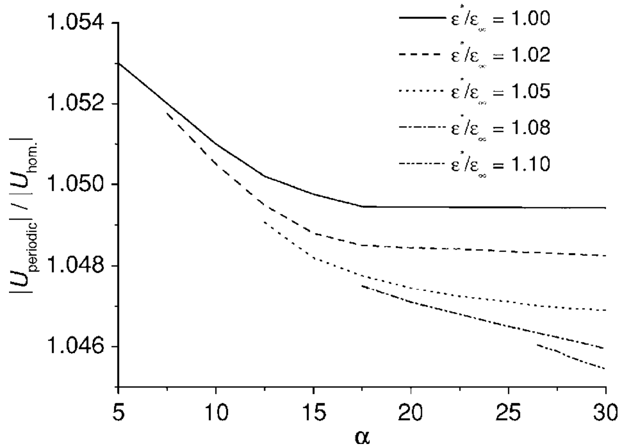

FIG. 8. The ratio $|U_{\text{periodic}}|/|U_{\text{hom}}|$ ($U<0$) of the potential energies of the periodic and homogeneous phases as a function of α for different values of $\varepsilon^*/\varepsilon_\infty$ (we fix $n=10^{19}$ cm^{-3}). The curves are referred to a temperature 5% below the critical one. All the curves but the solid one are limited on the left by the critical value of α.

V. CONCLUSIONS

We have studied in the framework of the Bogolyubov equilibrium distribution functions the transition of an uniform polaron system toward the formation of a periodic phase and its stability in time. Since the polaron distribution function $F_1(\boldsymbol{r})$ is a solution of a nonlinear integral equation of the Hammerstein type, we find the following.

(a) The periodic phase can appear because the integral equation shows the bifurcation phenomenon. We have calculated the highest transition temperature $T_{\text{cr}}^{(1)}$ as function of the polarity parameters of the medium $\varepsilon^*/\varepsilon_\infty$, of the electron-phonon coupling constant α and of the polaron density n. If we fix α and n, $T_{\text{cr}}^{(1)}$ is a decreasing function of $\varepsilon^*/\varepsilon_\infty$ [Fig. 2(a)], i.e., the less polar is the medium, the lower is $T_{\text{cr}}^{(1)}$. Moreover, a critical value of $(\varepsilon^*/\varepsilon_\infty)_{\text{cr}}$ appears, depending on α and n, such that for $\varepsilon^*/\varepsilon_\infty > (\varepsilon^*/\varepsilon_\infty)_{\text{cr}}$ the periodic phase cannot exist. On the other hand, if we fix $\varepsilon^*/\varepsilon_\infty$ and n, $T_{\text{cr}}^{(1)}$ is an increasing function of α, and a value α_{cr} exists such that only for $\alpha > \alpha_{\mathrm{cr}}$ the periodic phase appears [Fig. 2(b)]. α_{cr} is an increasing function of n (as a consequence of the collective effects) and of $\varepsilon^*/\varepsilon_\infty$ (Fig. 4). Finally, with fixed $\varepsilon^*/\varepsilon_\infty$ and α, it is found that $T_{\mathrm{cr}}^{(1)}$ increases with the density n (Fig. 5).

(b) The wave number k_{opt}, which is related to the distance between planes with the same value of the distribution function, has been calculated. If we fix n and the electron-phonon coupling constant α, k_{opt} is a slowly decreasing function of $\varepsilon^*/\varepsilon_\infty$ when the medium is very polar up to a specific value $(\varepsilon^*/\varepsilon_\infty)_1$, depending on α and n. For larger values of $\varepsilon^*/\varepsilon_\infty$, k_{opt} goes to zero very rapidly, indicating that the transition from the periodic to the uniform phase occurs nearly abruptly (Fig. 3).

(c) The kinetic stability of the periodic phase has been also studied. A lower bound $T_{\mathrm{cr}}^{(2)}$ on the temperature has been found for the periodic phase being stable in time. In other words, for temperatures lower than $T_{\mathrm{cr}}^{(2)}$, the periodic oscillation of the polaron gas are damped out, thus showing time instability. By combining this result with that in (a), we conclude that a temperature range $T_{\mathrm{cr}}^{(2)} < T < T_{\mathrm{cr}}^{(1)}$ exists, in which the periodic phase is stable. The temperature range $\Delta T = T_{\mathrm{cr}}^{(1)} - T_{\mathrm{cr}}^{(2)}$ decreases with the medium polarity, at fixed n and α [Fig. 6(a)]; this allows one to find $(\varepsilon^*/\varepsilon_\infty)_{\mathrm{cr}}$. Moreover, with fixed n and $\varepsilon^*/\varepsilon_\infty$, ΔT is an increasing function of α, that allows us to determine α_{cr} [Fig. 6(b)]. Finally we find that ΔT is an increasing function of n, if α and $\varepsilon^*/\varepsilon_\infty$ are fixed (inset of Fig. 5).

(d) The total energies of the periodic and homogeneous phases have been compared (Fig. 8), showing that if the above conditions of existence and stability are met, the periodic phase always is the lowest energy one.

At the moment, the only experimental indications of polaron structured states concern the formation of polaron clusters in high-T_c superconductors[9] and in manganites[6,8] with strong magnetic interactions. Until now there has been no specific indication for the formation of a periodic structure of polarons. This work indicates that the periodic structure can more likely form in strongly polar insulator or semiconductor materials with high electron-phonon coupling constant, large effective mass and with a small stationary number of electrons in the conduction band. Only halkali halides, such as LiF, or oxides, such as TiO$_2$, seem to be good candidates.

APPENDIX A

In this appendix we show the derivation of the solution of Eq. (20) where, as pointed out before, $\psi=0$ for small deviations from the homogeneous distribution of particles. If we define the operator $A=\boldsymbol{v}\cdot\boldsymbol{\nabla}_{\boldsymbol{r}}$, the solution of Eq. (20) can be given the following form:

$$\varphi(\boldsymbol{r},\boldsymbol{v},t) = \exp[-A(t-t_0)]\varphi(\boldsymbol{r},\boldsymbol{v},t_0) + \int_{t_0}^{t} \exp[-A(t-\tau)]\Psi(\boldsymbol{r},\boldsymbol{v},\tau)d\tau, \qquad \text{(A1)}$$

with

$$\Psi(\boldsymbol{r},\boldsymbol{v},t) = (1/M)\boldsymbol{\nabla}_{\boldsymbol{v}} f_0(\boldsymbol{v}^2)\cdot\boldsymbol{\nabla}_{\boldsymbol{r}}\int\int_{(\infty)}\Phi(|\boldsymbol{r}-\boldsymbol{r}'|) \times\varphi(\boldsymbol{r}',\boldsymbol{v}',t)d\boldsymbol{v}'d\boldsymbol{r}'.$$

The action of the operator $\exp[-A(t-t_0)]$ shifts the distribution function from the point with coordinates $(\boldsymbol{r},\boldsymbol{v},t)$ to the point $[\boldsymbol{r}-\boldsymbol{v}(t-t_0),\boldsymbol{v},t]$. Taking into account this property of the translation operator, we can rewrite the solution (A1) as follows

$$\varphi(\boldsymbol{r},\boldsymbol{v},t) = \varphi[\boldsymbol{r}-\boldsymbol{v}(t-t_0),\boldsymbol{v},t_0] + \int_{t_0}^{t} d\tau\frac{1}{M}\boldsymbol{\nabla}_{\boldsymbol{v}} f_0(\boldsymbol{v}^2) \cdot\boldsymbol{\nabla}_{\boldsymbol{r}}\int_{(\infty)}\Phi(|\boldsymbol{r}-\boldsymbol{v}(t-\tau)-\boldsymbol{r}'|)\rho(\boldsymbol{r}',\tau)d\boldsymbol{r}', \qquad \text{(A2)}$$

where $\rho(\boldsymbol{r},t)=\int_{(\infty)}\varphi(\boldsymbol{r},\boldsymbol{v},t)d\boldsymbol{v}$ is the spatial distribution of the particles. It can be obtained by integrating Eq. (A2) with respect to the velocity

$$\rho(\boldsymbol{r},t) = R(\boldsymbol{r},t,t_0) + \int_{(\infty)} d\boldsymbol{v}\int_{t_0}^{t} d\tau\frac{1}{M}\boldsymbol{\nabla}_{\boldsymbol{v}} f_0(\boldsymbol{v}^2) \cdot\boldsymbol{\nabla}_{\boldsymbol{r}}\int_{(\infty)}\Phi[|\boldsymbol{r}-\boldsymbol{v}(t-\tau)-\boldsymbol{r}'|]\rho(\boldsymbol{r}',\tau)d\boldsymbol{r}', \qquad \text{(A3)}$$

with $R(\boldsymbol{r},t,t_0)=\int_{(\infty)}\varphi[\boldsymbol{r}-\boldsymbol{v}(t-t_0),\boldsymbol{v},t_0]d\boldsymbol{v}$.

The solution $\rho(\boldsymbol{r},t)$ of Eq. (A3) can be found in the form $\rho(\boldsymbol{r},t)=\Sigma_k\rho_k(t)\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$. Furthermore (see Sec. III), we assume the initial condition $\varphi(\boldsymbol{r},\boldsymbol{v},t_0)=\Sigma_k\varphi_k(\boldsymbol{v},t_0)\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$, with $\varphi_k(\boldsymbol{v},t_0)=a_k f_0(\boldsymbol{v}^2)$. This gives the Volterra integral equation

$$\rho_k(t) = P_k(t-t_0) + \int_{t_0}^{t} Q_k(t-\tau)\rho_k(\tau)d\tau, \qquad \text{(A4)}$$

where

$$P_k(t-t_0) = a_k\int_{(\infty)} f_0(\boldsymbol{v}^2)\exp[-i\boldsymbol{k}\cdot\boldsymbol{v}(t-t_0)]d\boldsymbol{v},$$

$$Q_k(t-\tau) = \frac{i\sigma_0(k)}{M}\int_{(\infty)}\exp[i\boldsymbol{k}\cdot\boldsymbol{v}(t-\tau)]\boldsymbol{k}\cdot\boldsymbol{\nabla}_{\boldsymbol{v}} f_0(\boldsymbol{v}^2)d\boldsymbol{v},$$

$$\sigma_0(k) = \int_{(\infty)}\Phi[|\boldsymbol{r}-\boldsymbol{v}(t-\tau)-\boldsymbol{r}'|] \times\exp\{-i\boldsymbol{k}\cdot[\boldsymbol{r}-\boldsymbol{v}(t-\tau)-\boldsymbol{r}']d\boldsymbol{r}'\}.$$

Since the kernel depends on the difference $(t-\tau)$, the integral equation can be solved using the standard method of Laplace transform.[26] We obtain

$$\rho_k(t) = \frac{1}{2\pi i}\int_{g-i\infty}^{g+i\infty}\exp(pt)\frac{P_k(p)}{1-Q_k(p)}dp, \qquad \text{(A5)}$$

where $P_k(p)=\int_0^\infty\exp(-pt)P_k(t)dt$ and $Q_k(p)=\int_0^\infty\exp(-pt)Q_k(t)dt$. The integral (A5) is done in the complex plane of the variable p along a straight line parallel to the imaginary axis $(-g-i\infty,-g+i\infty)$. In order to obtain the solution in an analytical form, we approximate the Maxwell velocity distribution function with the symmetric function

$$\tilde{f}_0(v^2)=\frac{n\beta}{\pi}\frac{1}{1+v^2\beta^2},\quad \beta=[M/2\vartheta]^{1/2}, \tag{A6}$$

which is valid when the velocities are small, i.e., $(v\beta)\ll 1$. Using the approximation (A6) the kernel of the integral equation (A4) and the free term become

$$\begin{aligned}Q_k(t-\tau)&=\frac{ik\sigma_0(k)}{M}\int_{-\infty}^{+\infty}\exp[ikv(t-\tau)]\frac{d}{dv}\left(\frac{n\beta}{\pi}\frac{1}{1+v^2\beta^2}\right)dv\\&=\frac{k^2\sigma_0(k)n}{M}(t-\tau)\exp[-k(t-\tau)/\beta],\end{aligned}\tag{A7}$$

$$\begin{aligned}P_k(t)&=a_k\int_{-\infty}^{+\infty}\exp(-ikvt)\left(\frac{n\beta}{\pi}\frac{1}{1+v^2\beta^2}\right)dv\\&=a_kn\exp(-kt/\beta).\end{aligned}\tag{A8}$$

Equation (A8) clearly shows that the initial perturbation is produced by $\varphi_k(\boldsymbol{v},t_0)=a_kf_0(\boldsymbol{v}^2)$. Finally, it follows that

$$Q_k(p)=\int_0^\infty\exp(-pt)Q_k(t)dt=\frac{k^2\sigma_0(k)n}{M}\frac{1}{(p+k/\beta)^2},$$

$$P_k(p)=\int_0^\infty\exp(-pt)P_k(t)dt=\frac{a_kn}{(p+k/\beta)}\tag{A9}$$

and consequently the solution [Eq. (A5)] of the integral equation (A4) is given by

$$\begin{aligned}\rho_k(t)&=\frac{a_kn}{2\pi i}\int_{g-i\infty}^{g+i\infty}\exp(pt)\frac{p+k/\beta}{(p+k/\beta)^2-\Omega^2}dp\\&=a_kn\exp(-kt/\beta)\mathrm{ch}(\Omega t),\end{aligned}\tag{A10}$$

where $\Omega^2=-k^2\sigma_0(k)n/M$ is the square of a frequency. Equation (A10) is the same as Eq. (21).

APPENDIX B

In this appendix we show the derivation of the solution of Eq. (24). Using the relationship

$$\begin{aligned}\int_0^Z\exp(s^2)ds&=\left[\Omega\int_0^{(M/2\vartheta)^{1/2}/k}\exp[(\gamma^2-\Omega^2)x^2]\cos(2\gamma\Omega x^2)dx\right.\\&\quad\left.+\gamma\int_0^{(M/2\vartheta)^{1/2}/k}\exp[(\gamma^2-\Omega^2)x^2]\sin(2\gamma\Omega x^2)dx\right]\\&\quad+i\left[\gamma\int_0^{(M/2\vartheta)^{1/2}/k}\exp[(\gamma^2-\Omega^2)x^2]\cos(2\gamma\Omega x^2)dx\right.\\&\quad\left.-\Omega\int_0^{(M/2\vartheta)^{1/2}/k}\exp[(\gamma^2-\Omega^2)x^2]\sin(2\gamma\Omega x^2)dx\right],\end{aligned}\tag{B1}$$

Eq. (24) can be rewritten as

$$\begin{aligned}\frac{\vartheta}{n\sigma_0(k)}&=-1+\frac{2}{k}\left(\frac{M}{2\vartheta}\right)^{1/2}\cos\left(\frac{M\gamma\Omega}{\vartheta k^2}\right)\exp\left(-\frac{M}{2\vartheta k^2}\right)\\&\quad\times[(\Omega^2-\gamma^2)I_1-2\gamma\Omega I_2]+\frac{2}{k}\left(\frac{M}{2\vartheta}\right)^{1/2}\sin\left(\frac{M\gamma\Omega}{\vartheta k^2}\right)\\&\quad\times\exp\left(-\frac{M}{2\vartheta k^2}\right)[(\Omega^2-\gamma^2)I_2+2\gamma\Omega I_1]\\&\quad+i\frac{2}{k}\left(\frac{M}{2\vartheta}\right)^{1/2}\cos\left(\frac{M\gamma\Omega}{\vartheta k^2}\right)\exp\left(-\frac{M}{2\vartheta k^2}\right)\\&\quad\times[(\Omega^2-\gamma^2)I_2+2\gamma\Omega I_1]+i\frac{2}{k}\left(\frac{M}{2\vartheta}\right)^{1/2}\sin\left(\frac{M\gamma\Omega}{\vartheta k^2}\right)\\&\quad\times\exp\left(-\frac{M}{2\vartheta k^2}\right)[(\Omega^2-\gamma^2)I_1+2\gamma\Omega I_2],\end{aligned}\tag{B2}$$

with

$$I_1=\int_0^{(M/2\vartheta)^{1/2}/k}\exp[-(\gamma^2-\Omega^2)x^2]\cos(2\gamma\Omega x^2)dx,$$

$$I_2=\int_0^{(M/2\vartheta)^{1/2}/k}\exp[(\gamma^2-\Omega^2)x^2]\sin(2\gamma\Omega x^2)dx.$$

Equation (B2) allows us to determine, for a given polaron density n and polarity parameters of the material, the damping parameter γ. The stability region of the periodic polaron phase if found for temperatures such that Eq. (B2) has a purely imaginary solution γ.

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