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On the polaron mass at finite temperatures

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Abstract. The concept of polaron mass at $T \neq 0$ is shown to be poorly defined. Various forms of temperature dependence $m_{eff}(T)$ suggested in the literature do not coincide, their difference being due to the different ways of defining the mass at $T \neq 0$. A definition of effective mass as the ratio of the impulse $F\,dt$ and the mean polaron velocity $\langle v \rangle_T$ is suggested and the elementary derivation of the temperature dependence of the mass spectrum in such a way in the region $kT \ll \hbar\omega_{ph}$ is given. The results obtained for the dependence $m_{eff}(T)$ via polaron cyclotron resonance frequency are discussed and the width of the cyclotron line is shown to be of the same order as or larger than the temperature shift of its maximum, the latter being not given by any of the expressions for $m_{eff}(T)$ discussed earlier. We give an expression for the shape of the resonance line and compare it with the available data.

1. Introduction

The Fröhlich polaron, i.e. the electron interacting with long-wave optical phonons in the polar crystal, is known as one of the most simple and most beautiful problems of solid state physics where the methods of quantum field theory proved to be very efficient and the quantitative comparison of the theoretical predictions with experiment is possible [1, 2]. One of the main characteristics of a polaron is its effective mass $m_{eff}(T)$. There are different ways to calculate this value. At zero temperature all methods give identical answers coinciding with experiment. At finite temperatures the situation is much less clear. Different ways to define $m_{eff}(T)$ at $T \neq 0$ have been used: via the mean energy of the state with momentum p [3], as the pole of the temperature-dependent Green function [4, 5], via the interaction-induced contribution in the free energy of the system [6] and via the Feynman path integral [7–9]. The temperature dependences of all these different 'masses' are by no means the same although at the limit $T = 0$ all these definitions coincide. Note, however, that in the low-temperature region the most popular (at present) definitions in [6] and in [7–9] coincide even at $T \neq 0$.

The question arises of whether the temperature dependence of the polaron mass can be measured experimentally and compared with conflicting theoretical predictions so that one could decide which of these different definitions of $m_{eff}(T)$ should be preferred to others.

Unfortunately, as we shall see, the answer to this question is negative and neither of these different definitions is connected *directly* to physically measurable quantities (the definition used in [6–9] is, however, connected *indirectly* with the mobility of polarons and, if handled with care, should perhaps be preferred to others).

This work consists of two simple considerations. First, we give an elementary interpretation of the temperature dependence of the effective mass defined in [6–9] at $T \ll \omega_{ph}$, where ω_{ph} is the frequency of optical phonons (here and in the following we use the convention $\hbar = c = k = 1$). Second, we show that in cyclotron resonance experiments quite a different 'mass' is measured and, further, the width of the cyclotron line at $T \neq 0$ is always of the same order as or much greater than the temperature shift of its maximum. Thus at $T \neq 0$ the concept of mass is not universal and it is better not to introduce it at all.

2. 'Inertial' polaron mass

The Fröhlich Hamiltonian describing the interaction of the electron with the polar crystal lattice has the form [1]

$$H = \frac{p^2}{2m^*} + \omega_{ph} \int \frac{dk}{(2\pi)^3} a_k^\dagger a_k + i(2\pi\sqrt{2}\alpha)^{1/2} \left(\frac{\omega_{ph}^3}{m^*}\right)^{1/4} \int \frac{dk}{(2\pi)^3} \frac{1}{|k|}$$
$$\times (a_k^*) \exp(-ik \cdot x) - a_k \exp(ik \cdot x) \qquad (1)$$

where $a_k^\dagger$ and a_k are the phonon creation and annihilation operators and m^* is the effective electron mass without taking into account its interaction with long-wave modes. The interaction with the lattice leads to renormalization of the polaron dispersion law. In the first order of the coupling constant which we assume to be small it has the form [1, 2]

$$E(p) = p^2/2m^* - \alpha\omega_{ph} (\sqrt{2\omega_{ph} m^*}/p) \sin^{-1} (p/\sqrt{2\omega_{ph} m^*}). \qquad (2)$$

At $p > \sqrt{2\omega_{ph} m^*}$, $E(p)$ acquires an imaginary part which corresponds to instability of the fast-moving polarons with respect to phonon emission. However, in the low-temperature region $T \ll \omega_{ph}$ the fraction of such fast polarons as well as the density of real optical phonons in the crystal are exponentially small. Thus at $T \ll \omega_{ph}$ the presence of the latter may be safely neglected and we have simply the Boltzmann distribution of charged particles:

$$f(p) \propto \exp[-E(p)/T] \qquad (3)$$

where $E(p)$ is the renormalized dispersion law (2).

Let us define the temperature-dependent polaron 'inertial' mass as

$$m_{eff}(T) = F\, dt/\langle v \rangle_T \qquad (4)$$

where $\langle v \rangle_T$ is the mean polaron velocity acquired under the action of the infinitesimal impulse $F\, dt$. $\langle v \rangle_T$ can be expressed in the following way:

$$\langle v \rangle_T = \int dp \frac{\partial E(p)}{\partial p} \exp[-E(|p - F\, dt|)/T] \bigg/ \int dp \exp[-E(p)/T]. \qquad (5)$$

Substituting $E(p)$ here in the form (2), expanding the numerator over $F\, dt$ and the whole expression over the small coupling constant α, we get

$$m_{eff}(T) = m^*\{1 + (\alpha\sqrt{\pi}/3)(\omega_{ph}/T)^{5/2} \exp(-\omega_{ph}/T) [(1 - \omega_{ph}/T) I_0(\omega_{ph}/2T)$$
$$- I_1(\omega_{ph}/2T)] + O[\exp(-\omega_{ph}/T)]\} \qquad (6)$$

(I_0 and I_1 are the modified Bessel functions). This expression coincides with equation (3.8) of [9], is in accordance with the results of [6–8] but differs from effective temperature-dependent polaron masses defined in [3–5]. The low-temperature expansion of equation (6) has the form

$$m_{\rm eff}(T) = m^*[1+(\alpha/6)(1+9T/4\omega_{\rm ph}+225T^2/32\omega_{\rm ph}^2+\ldots)] \qquad (7)$$

i.e. the effective mass increases with increasing temperature. This has a clear physical interpretation: *the dispersion law (2) is non-parabolic and the effective mass of the* polaron with momentum p, given by

$$m_{\rm eff}(T) = p/(\partial E/\partial p) \qquad (8)$$

increases with increasing p and the mass averaged over the distribution (3) increases with increasing T (see [6]).

3. Cyclotron resonance on polarons

The effective mass defined according to equation (4) is related to the problem of polaron mobility at $T \neq 0$ [7–10]. However, the polaron mobility depends crucially also on the complicated processes of polaron scattering by phonons and impurities and the direct measurement of the value (4) is not possible. The best way to measure polaron mass is by the cyclotron resonance experiment.

At $T = 0$ the polaron momentum is equal to zero and the cyclotron frequency is $\omega_c = eH/m_{\rm eff}(0)$. At $T \neq 0$ the non-parabolic dispersion law (20) results in different cyclotron frequencies of polarons with different momenta (the momentum distribution being given by equation (3)) and the cyclotron resonance line acquires a width, its maximum being shifted at lower frequencies. Such a widening of the cyclotron line is well known in relativistic plasmas [11]. In the polaron problem the widening of the cyclotron line due to non-parabolicity of the spectrum was noted in [12] but no quantitative estimate has been made. Let us find the shape of cyclotron line at $T \ll \omega_{\rm ph}$. The effects due to scattering by phonons and impurities will be neglected.

At small T the typical polaron momenta are small and the way in which the small deviations from parabolicity in the dispersion law (2)

$$E(p) = \text{constant} + (p^2/2m^*)(1-\alpha/6) - 3\alpha p^4/160m^{*2}\omega_{\rm ph} - O(p^6) \qquad (9)$$

are taken into account is sufficient. We restrict ourselves to the case of a weak magnetic field ($\omega_c \ll T \ll \omega_{\rm ph}$) where the classical treatment is adequate. In this case the effective Hamiltonian of a polaron in the external magnetic field is dictated by gauge invariance considerations: $\mathcal{H}(p) = E(\mathcal{P})$ where $\mathcal{P} = \sqrt{|p-eA|^2}$. Writing down and solving the Hamiltonian equations of motion, we may easily find the Larmor frequency of the polaron in an external magnetic field as the function of polaron momentum p:

$$\omega(p) = 2eH\,\partial E/\partial p^2 = (eH/m^*)(1-\alpha/6-3\alpha p^2/40m^*\omega_{\rm ph}+\ldots). \qquad (10)$$

The power absorption in the frequency interval $d\omega$ is

$$f(\omega)\,d\omega \propto \exp[-E(p)/T]\,|p_\perp|\,dp \propto p^2\exp(-p^2/2m^*)\,(dp^2/d\omega)\,d\omega$$

and hence

$$f(\omega) \propto (\omega_c-\omega)\exp[20(\omega-\omega_c)\omega_{\rm ph}/3\alpha\omega_c T]\,\theta(\omega_c-\omega) \qquad (11)$$

where $\omega_c = (eH/m^*)(1-\alpha/6)$ is the cyclotron frequency at $T = 0$. The finite-temperature cyclotron line (11) has a maximum at

$$\omega = \omega_c(1-3\alpha T/20\omega_{\rm ph}+C\alpha T^2/\omega_{\rm ph}^2+\ldots). \qquad (12)$$

The decrease in the resonance maximum frequency at non-zero T can be interpreted

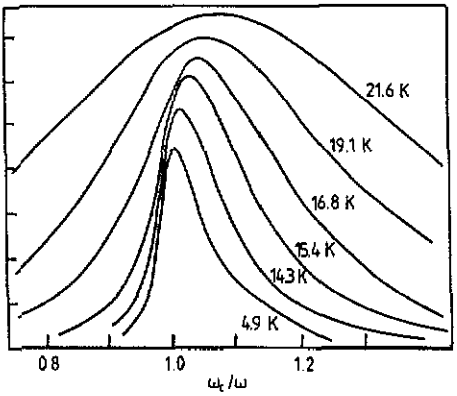

Figure 1. The shape of the cyclotron resonance lines at different temperatures as measured in [13]. The power absorption in arbitrary units is plotted along the vertical axis.

as the growth of the effective 'cyclotron mass'. Note, however, that such a 'cyclotron mass' does not coincide with the 'inertial mass' (4.7) discussed earlier. This difference seems quite natural as the width of 'mass distribution' (11) is comparable with the shift of its maximum.

Turning to the question of comparison of our results (11) and (12) with experiment we meet immediately two important difficulties. First of all in real experimental situations the cyclotron resonance width is usually provided not by non-parabolicity of the polaron spectrum but rather by the processes of polaron scattering by acoustic phonons and impurities (if $T \ll \omega_{ph}$, the density of optical phonons is exponentially small). The second and most troublesome difficulty is that, besides the 'long-wave' non-parabolicity considered, the spectrum also involves the intrinsic 'short-wave' non-parabolicity due to the terms proportional to p^4, etc, which enter directly the Fröhlich Hamiltonian (1). The effects due to intrinsic non-parabolicity have exactly the same form and temperature dependence as the effects due to optical phonon interaction and we do not see any way to distinguish between them.

The cyclotron resonance of polarons at low temperatures was studied experimentally in [13]. The study has been performed for the polarons in the ionic crystal AgBr with the coupling constant $\alpha \simeq 1.6$ in the temperature region from 5 to 22 K which is much less than $\omega_{ph} \simeq 200$ K (the parameters of the Fröhlich Hamiltonian for AgBr can be found for example in [1]). The coupling constant $\alpha = 1.6$ may be considered as 'small' as the higher-order effects become essential at $\alpha \simeq 5$–6 (see, e.g., [14]).

The shape of the cyclotron resonance line measured in [13] is presented in figure 1. We see immediately that at $T > 17$ K the line is rather broad and its shape is symmetrical and described adequately by the Lorentzian

$$f(\omega) \propto 1/[1 + (\omega - \omega_c)^2 \tau^2] \tag{13}$$

where τ is the relaxation time due to polaron scattering processes. The physical mechanism of this scattering is, of course, an interesting but separate question (perhaps, the trap model discussed in [13] is adequate). At lower temperatures $T < 15$ K the line is much narrower, sharply asymmetrical and rather well approximated by the dependence

$$f(\omega) \propto \Delta\omega \exp[-\Delta\omega/C(T)]\, \theta(\Delta\omega)$$

following from equation (11). The step function dependence proportional to $\Delta\omega\ \theta(\Delta\omega)$

is somewhat smeared out, of course, but at low temperatures the smearing out is rather weak. We may conclude that in the low-temperature region the scattering effects are not very important. This conforms to theoretical expectations: at low temperatures the density of acoustic phonons falls off as T^3 and the resonance trap scattering discussed in [13] falls off exponentially proportionally to $\exp(-E_b/T)$ (E_b is the trap bound-state energy).

Unfortunately, the linewidth obtained from experimental data is several times larger than that obtained from our result (11). This means that the effects due to intrinsic non-parabolicity of the band are *more* important than those due to optical phonon interactions. One can see further that the linewidth *is not* proportional to T as follows from our simple classical theory based on the non-parabolicity of the spectrum and the experimental temperature dependence of the width is weaker.

Note that at the lowest temperature at which the measurements in [13] were done ($T_{min} = 4.9$ K) we are not, strictly speaking, in the classical region. The resonance frequency of [13] was about $\nu_c \simeq 5.5$ GHz which corresponds to $T_c \simeq 3$ K. It is not very much smaller than T_{min} and quantum corrections to our classical theory may be essential. However, quantum effects result on the contrary in the temperature dependence of the resonance line width being sharper than the classical theory predicts; at $T \ll T_c$ the width is exponentially small.

As for the shift of the resonance line maximum position, it is not seen at all at $T \leqslant 10$ K. The results for the shift plotted in figure 1 of [13] are consistent both with zero and with the weak temperature dependence of equation (12). Looking at the curves for the resonance lineshape at different temperatures 'with a magnifying glass', one may still guess that the maximum is shifted.

However, there are no experimental errors in the figure for the resonance line shapes quoted in [13] and it is not quite clear whether it is appropriate to analyse these curves quantitatively in too much detail. Further measurements of the resonance lineshapes in the low-temperature region on clean samples are required in order to check the prediction of equation (11) for the lineshape (with some additional numerical factor accounting for the intrinsic non-parabolicity effects present in the exponent).

In conclusion we repeat our main message: the polaron mass at $T \neq 0$ is a poorly defined concept and is not especially physical. At any rate, to compare the experimental shift of the cyclotron line maximum with the temperature dependence of the 'inertial mass' (7) as was done in [8] is rather pointless.

The difficulties of defining mass at non-zero temperature are rather universal and not special at all to polarons; see, e.g., our work [15] where a similar problem is discussed for solitons in relativistic field theory at non-zero temperatures.

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