

phys. stat. sol. (b) **109**, 599 (1982)

Subject classification: 6 and 13.4; 20.3; 22

*Sektion Physik der Humboldt-Universität zu Berlin,  
Bereich Theoretische Halbleiterphysik<sup>1)</sup> und Bereich Halbleiteroptik<sup>2)</sup>*

## Theory of Non-Radiative Multiphonon Capture Processes

### Solution of Old Controversies

By

K. PEUKER, R. ENDERLEIN, A. SCHENK, and E. GUTSCHE

The solution is given for long persisting controversial problems in the theory of non-radiative multiphonon capture of free carriers. Taking the adiabatic wave functions as basic states for the perturbational treatment of the transition probability particular attention is focused on the correct determination of the transition matrix elements. In this framework it is shown that to first order in the non-diagonal matrix element of the electron-lattice interaction all approaches developed so far (Condon approximation, non-Condon approximation, static approximation, etc.) lead to the same result, in contradiction to statements in the literature. In particular, the reason is found why earlier results for the transition rate within the adiabatic and Condon approximation are much too small. The static approximation is shown to be the lowest approximation to the adiabatic approach.

In der vorliegenden Arbeit werden über mehrere Jahrzehnte andauernde Kontroversen in der Theorie strahlungsloser Vielphononen-Prozesse gelöst. Bei der störungstheoretischen Behandlung der Übergangswahrscheinlichkeit auf der Grundlage adiabatischer Basiszustände wird der korrekten Berechnung der Übergangsmatrixelemente besondere Aufmerksamkeit gewidmet. Im Gegensatz zu Aussagen in der Literatur wird gezeigt, daß alle bisher bekannten Zugänge (Condon-Näherung, Nicht-Condonsche Näherung, Statische Näherung usw.) in niedrigster Ordnung bezüglich der nichtdiagonalen Elektron-Gitter-Kopplung zu dem gleichen Resultat führen. Insbesondere wird geklärt, warum in früheren Arbeiten im Rahmen der adiabatischen und Condon-Näherung viel zu kleine Werte für die Übergangswahrscheinlichkeit erhalten wurden. Die statische Näherung erweist sich als die niedrigste Approximation für die adiabatische Theorie.

### 1. Introduction

The theory of non-radiative multiphonon capture of free charge carriers belongs to the very few fields of solid state physics where fundamental controversies have been persisting over several decades of years, and where a satisfactory solution has not been achieved so far. The idea of the simultaneous emission of many phonons by an electron-hole pair which recombines across the gap was first introduced by Frenkel [1] and Möglich and Rompe [2]. The latter authors gave the first theoretical description by applying Bloch's approach to electron-lattice coupling in higher-order perturbation theory. Multiphonon transitions of carriers into localized defect states within the gap were treated theoretically for the first time in two independent papers, one by Huang and Rhys [3] and the other by Kubo [4]. These pioneering papers, although being related to different systems and limiting cases, were based on the same fundamental concept. The stationary states of carriers before and after trapping were taken in the adiabatic approximation [5]. The multiphonon capture processes were

<sup>1)</sup> August-Bebel-Platz, DDR-1080, GDR.

<sup>2)</sup> Neue Schönhauser Str. 20, DDR-1020 Berlin, GDR.

considered to be due to the non-adiabatic parts of the full electron-lattice Hamiltonian, and the non-adiabatic interaction terms were treated in first-order perturbation theory. Remember that the adiabatic approximation allows the separate determination of electronic and lattice wave functions, the former ones depending on lattice coordinates and the latter ones on electron states. Even if the electron-lattice interaction is taken in the linear approximation as has been done for the electronic wave functions in [3, 4], the lattice wave functions for different electron states are no longer orthogonal to each other and in such a way multiphonon transitions become possible. The linear approximation for the electronic wave functions with respect to the electron-lattice interaction leads to the independence of the electronic transition matrix element on lattice coordinates, an approximation which commonly is referred to as Condon approximation in analogy to the theory of optical transitions. Following Huang and Rhys [3] and Kubo [4] the adiabatic concept and the Condon approximation were employed in several other papers, among them those by Krivoglaz [6], Lax [7], and Kubo and Toyozawa [8]. More exact electronic wave functions with respect to the electron-lattice interaction were considered by Kovarskii [9]. Whereas the same exponential temperature dependence in the high-temperature region was obtained by Kovarskii, the magnitude of the pre-exponential factor of the capture cross-section turned out to be by a factor  $(\Delta E/\hbar\omega_0)^2$  larger than that of the Condon approximation ( $\Delta E$  is the electronic transition energy and  $\hbar\omega_0$  the phonon energy). Since this result was quite unexpected and also since it was derived by a rather difficult calculation it had been completely overlooked for many years. In recent years, calculations within the "non-Condon" approach were carried out by Ridley [10] and Goto et al. [11]. Other authors, in particular, Helmis [12], Kovarskii et al. [13], Haug [14], and Pässler [15 to 20], used instead of the adiabatic the so-called static approach [21]. In this approach the electronic wave functions are independent of the lattice coordinates whereas the lattice wave functions depend still on the electronic states. Pässler interpreted his static calculations as to be entirely non-equivalent to the adiabatic approach. This interpretation seemed to be supported by the fact that the static results differed from the adiabatic results within the Condon approximation [3, 4] by orders of magnitude. However, as it has to be expected and as will be shown explicitly in this paper, the capture cross-section derived within the static approach is the lowest-order approximation to the adiabatic result if the latter one is derived rigorously.

Meanwhile experimental results on multiphonon capture cross-sections from DLTS measurements by Henry, Lang, and Logan [22, 23] suggested that the earlier results in the adiabatic and Condon approximation for the absolute value of the capture cross-section were much too small and the much larger values by Kovarskii, Haug and Pässler were more adequate. Henry and Lang [23] tried to reproduce the order of magnitude observed experimentally by combining the adiabatic theory with the theory by Landau [24] and Zener [25] for the predissociation of molecules. However, as far as transition probabilities are calculated only in first-order perturbation theory, the Landau-Zener approach is essentially the quasi-classical limit of the adiabatic theory. In [23], practically, the results of the static limit of the adiabatic theory were reproduced. The Landau-Zener theory was exploited by Henry and Lang [23] for the treatment of reemission processes of carriers after capture. In a more accurate manner the Landau-Zener theory was applied to this problem by Sumi [26].

This survey of the literature shows that there are two different groups of papers on multiphonon capture of free carriers with regard to the absolute values of the capture cross-section. The first group [3, 4, 6 to 8] gave relatively small values and the second group [9 to 20] gave values by orders of magnitude larger which better agree with experimental observations. The question arises why these two groups of papers give

such different results although they appear to be only different versions of essentially the same order of perturbation theory. Another question is why the second group of papers give more adequate results than the first one. Finally, one may ask why the completely different approaches within the second group — non-Condon and static — result in the same order of magnitude.

These questions will be answered in the present paper. Partial answers have already been given by other authors. For the case of a particular choice of non-Condon adiabatic wave functions the equivalence between the adiabatic and static approaches was noticed by Kovarskii et al. [13]. Quite recently, Huang [27] and one of the present authors (E.G.) [28, 29] used non-Condon wave functions similar to those by Kovarskii and others [9, 10, 30, 31] to demonstrate this equivalence in a much more direct and explicit way. In addition, Huang gave an argument why non-Condon adiabatic wave functions should be used to avoid inconsistencies within the adiabatic theory.

As we will show, however, Condon functions can be applied as well if the adiabatic theory is treated rigorously. This means, in particular, that for the time development of the quasi-stationary states the full non-diagonal part of the total Hamiltonian has to be taken into account rather than only its non-adiabatic contribution, as has been done so far in all papers based on the adiabatic approach. In this paper we will show why and how the adiabatic theory of non-radiative multiphonon transitions has to be modified to obtain a unified picture where no discrepancies are left between the Condon and non-Condon approximations and also between these approximations and the static results.

## 2. Transition Matrix Elements

As customary, we start from an effective Hamiltonian for the electron–lattice system of the form

$$H(x, Q) = H_e(x) + V(x, Q) + H_L(Q), \quad (1)$$

where  $x$  are electronic coordinates and  $Q$  are normal coordinates of vibration. The term  $H_e(x)$  refers to the electronic subsystem. In a single mode model and in terms of non-dimensional normal coordinates the Hamiltonian  $H_L(Q)$  of the free lattice is given by

$$H_L(Q) = \frac{1}{2} \hbar \omega_0 \left( -\frac{\partial^2}{\partial Q^2} + Q^2 \right). \quad (2)$$

The electron–lattice coupling reads in the linear approximation

$$V(x, Q) = v(x) Q. \quad (3)$$

The total wave function  $\Psi(x, Q)$  satisfying the Schrödinger equation  $H\Psi = E\Psi$  is taken in the form

$$\Psi(x, Q) = \varphi(x, Q) \Phi(Q). \quad (4)$$

The two functions  $\varphi(x, Q)$  and  $\Phi(Q)$  are chosen as the solutions of the coupled set of equations [32]

$$(H_e(x) + V(x, Q)) \varphi(x, Q) + \Phi^{-1}(Q) [H_L(Q), \varphi(x, Q)] \Phi(Q) = U(Q) \varphi(x, Q), \quad (5)$$

$$(H_L(Q) + U(Q)) = E\Phi(Q), \quad (5a)$$

where the square brackets  $[.,.]$  stand for the commutator. Within the adiabatic approximation the coupling term in (5) is neglected by setting the operator of the non-

adiabaticity

$$L(\varphi\Phi) = [H_L, \varphi] \Phi \quad (6)$$

formally equal to zero. One gets

$$(H_e(x) + V(x, Q)) \varphi_n(x, Q) = U_n(Q) \varphi_n(x, Q), \quad (7)$$

$$(H_L(Q) + U_n(Q)) \Phi_{nN}(Q) = E_{nN} \Phi_{nN}(Q) \quad (7a)$$

with  $\varphi_n(x, Q)$  and  $\Phi_{nN}(Q)$  as the adiabatic wave functions. Now the assumption is made that the quantum states of an electron in a deformable crystal lattice can be identified with the adiabatic wave functions or with certain reasonable approximations to them. The time evolution of the exact or approximate adiabatic functions is governed by the total Hamiltonian (1). The transition probability per unit time between different electronic terms may be calculated by the "golden rule"

$$W_{nn'} = \frac{2\pi}{\hbar} \sum_{NN'} p_N |(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'})|^2 \delta(E_{nN} - E_{n'N'}), \quad (8)$$

where  $p_N$  is the statistical factor for the initial lattice states.

To establish the relationship between the different approaches to the theory of non-radiative multiphonon capture processes we concentrate our attention to the transition-inducing non-diagonal matrix elements of the full Hamiltonian  $(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'})$ . Provided that the wave functions  $\varphi_n(x, Q)$   $\Phi_{nN}(Q)$  are exact solutions of the adiabatic equations (7) and (7a) it is easy to show that the non-diagonal matrix elements of the full Hamiltonian  $H$  can be expressed by (cf. [3])

$$(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'}) = (\Phi_{nN}\varphi_n | L | \varphi_{n'}\Phi_{n'N'}). \quad (9)$$

Relation (9) has been exclusively used in all calculations of the transition rate on the basis of the adiabatic approximation. However, it has been overlooked that in the case of an approximate solution of (7) and (7a) relation (9) has to be modified according to<sup>3)</sup>

$$(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'}) = A_{nNn'N'} + B_{nNn'N'}, \quad (10)$$

$$A_{nNn'N'} = (\Phi_{nN}\varphi_n | H_e + V | \varphi_{n'}\Phi_{n'N'}), \quad (10a)$$

$$B_{nNn'N'} = (\Phi_{nN}\varphi_n | L | \varphi_{n'}\Phi_{n'N'}). \quad (10b)$$

The first term  $A_{nNn'N'}$  on the right-hand side of (10) vanishes if the  $\varphi_n$  satisfy equation (7) exactly. But commonly perturbative solutions with respect to  $V(x, Q)$  are used for  $\varphi_n$ . In this case the expression  $A_{nNn'N'}$  can play an important role, as we will see later. Only by including this term one can derive correct results for the transition rate.

We now proceed to investigate in a systematic way the perturbative solutions of (7) and (7a) with respect to the electron lattice coupling  $V(x, Q)$ . To zeroth order we have

$$\varphi_n(x, Q) = \varphi_n^{(0)}(x), \quad U_n(Q) = U_n^{(0)} \quad (11)$$

and the vibration functions  $\Phi_{nN}^{(0)}(Q)$  (9) are determined from the equation

$$(H_L^{(0)}(Q) + U_n^{(0)}) \Phi_{nN}^{(0)}(Q) = E_{nN}^{(0)} \Phi_{nN}^{(0)}(Q). \quad (12)$$

<sup>3)</sup> Throughout the paper only non-diagonal transition matrix elements are considered, i.e.  $n \neq n'$  everywhere.

Since  $U_n^{(0)}$  does not depend on  $Q$  the zeroth-order approximation is not associated with any  $Q$ -dependent potential differences in the vibrational equations belonging to different electronic terms, and is therefore not capable to describe multiphonon but only one-phonon transitions. The corresponding matrix elements are

$$(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'}) = v_{nn'}^{(0)}(\Phi_{nN}^{(0)} | Q | \Phi_{n'N'}^{(0)}), \quad (13)$$

$$v_{nn'}^{(0)} = (\varphi_n^{(0)} | v | \varphi_{n'}^{(0)}). \quad (13a)$$

A better solution of equation (7) is provided by the first-order or Condon expression

$$\varphi_n^C(x, Q) = \varphi_n^{(0)}(x) + \sum_m' \frac{v_{mn}^{(0)}Q}{U_n^{(0)} - U_m^{(0)}} \varphi_m^{(0)}(x) \quad (14)$$

with the corresponding vibrational equation

$$(H_L(Q) + U_n^{(0)} + v_{nn}^{(0)}Q) \Phi_{nN}^{(1)}(Q) = E_{nN}^{(1)}\Phi_{nN}^{(1)}(Q). \quad (15)$$

The linear term  $v_{nn}^{(0)}Q$  in (15) results in shifts of the equilibrium positions of the normal modes and, thus, yields the simplest model for the description of multiphonon processes. The transition matrix elements become

$$A_{nNn'N'}^C = v_{nn'}^{(0)} \frac{v_{n'n'}^{(0)} - v_{nn}^{(0)}}{U_n^{(0)} - U_{n'}^{(0)}} (\Phi_{nN}^{(1)} | Q^2 | \Phi_{n'N'}^{(1)}) + O(v_{nn'}^{(0)2}), \quad (16)$$

$$B_{nNn'N'}^C = \frac{v_{nn'}^{(0)}}{U_{n'}^{(0)} - U_n^{(0)}} (\Phi_{nN}^{(1)} | [H_L, Q] | \Phi_{n'N'}^{(1)}) + O(v_{nn'}^{(0)2}). \quad (17)$$

At this stage of the development of the adiabatic theory one can link it with the static approach. As employed in the literature, the static approximation means the use of zeroth-order electronic wave functions from (11) and of first-order lattice functions from (15). With this modification the transition matrix element  $B_{nNn'N'}$  of the non-adiabaticity operator vanishes exactly, and the transition operator coincides with the electron-lattice interaction potential. Then the total transition matrix element has the form (13), but with the first-order lattice function  $\Phi_{nN}^{(1)}$  instead of the zeroth-order function,

$$(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'}) = v_{nn'}^{(0)}(\Phi_{nN}^{(1)} | Q | \Phi_{n'N'}^{(1)}) \quad (18)$$

with  $v_{nn'}^{(0)}$  from (13a).

In all earlier papers, based on the adiabatic concept, instead of the total sum (10) only the matrix elements  $B_{nNn'N'}$  (10b) were taken into account. Especially, in the Condon approximation (14) and (15) from the equivalent form

$$(\Phi_{nN}^{(1)} | [H_L, Q] | \Phi_{n'N'}^{(1)}) = -\hbar\omega_0 \left( \Phi_{nN}^{(1)} \left| \frac{\partial}{\partial Q} \right| \Phi_{n'N'}^{(1)} \right) \quad (19)$$

the conclusion was drawn that this approximation yields a transition rate smaller than that of the static approximation (11) by the factor  $(\hbar\omega_0/(U_n^{(0)} - U_{n'}^{(0)}))^2$ . The error of this conclusion can be seen by using an alternative transformation<sup>4)</sup> of  $B_{nNn'N'}$  based

<sup>4)</sup> See also Huang [27] and Gutsche [28, 29].

on the equation of motion (15), namely

$$\begin{aligned} (\Phi_{nN}^{(1)} | [H_L, Q] | \Phi_{n'N'}^{(1)}) &= (E_{nN} - E_{n'N'} - (U_n^{(0)} - U_{n'}^{(0)})) (\Phi_{nN}^{(1)} | Q | \Phi_{n'N'}^{(1)}) - \\ &- (v_{nn}^{(0)} - v_{n'n'}^{(0)}) (\Phi_{nN}^{(1)} | Q^2 | \Phi_{n'N'}^{(1)}). \end{aligned} \quad (20)$$

Inserting (20) into (17), we obtain by observing the conservation of energy ( $E_{nN} = E_{n'N'}$ ) the final result for the sum

$$A_{nNn'N'}^C + B_{nNn'N'}^C = v_{nn'}^{(0)} (\Phi_{nN}^{(1)} | Q | \Phi_{n'N'}^{(1)}) + O(v_{nn'}^{(0)2}), \quad (21)$$

which is approximately the same result as in the static approximation according to (18). The difference between (21) and (18) is quadratic in the non-diagonal matrix elements  $v_{nn'}^{(0)}$  and usually can be neglected.

We now proceed to the non-Condon approximation developed by Kovarskii and coworkers [9, 30, 31]. In this case the electronic wave functions are approximated by the first-order expression according to Brillouin-Wigner perturbation theory<sup>5</sup>)

$$\varphi_n^{NC}(x, Q) = \varphi_n^{(0)}(x) + \sum'_m \frac{v_{mn}^{(0)} Q}{\tilde{U}_n(Q) - \tilde{U}_m(Q)} \varphi_m^{(0)}(x), \quad (22)$$

$$\tilde{U}_n(Q) = U_n^{(0)} + v_{nn}^{(0)} Q. \quad (22a)$$

The vibrational equation is the same apart from higher-order terms as in the Condon case (15) and is, therefore, still harmonic. In contrast to the foregoing case the main contribution is now provided by the matrix element  $B_{nNn'N'}^{NC}$  which is associated with the non-adiabaticity operator, whereas the magnitude of  $A_{nNn'N'}^{NC}$  is of the order  $v_{nn'}^{(0)2}$ , as can be easily derived. The analytical complications of the non-Condon approach

Table 1

Contributions  $A_{nNn'N'}$  and  $B_{nNn'N'}$  to the transition matrix element  $(\Phi_{nN}\varphi_n | H | \varphi_{n'}\Phi_{n'N'})$  linear in  $v_{nn'}^{(0)}$  within different approximations. Note that in all cases  $A_{nNn'N'} + B_{nNn'N'}$  has the same value indicating that the different approximations used in the theory of multiphonon capture are identical to first order in the electron-lattice interaction

	$A_{nNn'N'}$	$B_{nNn'N'}$	$(\Phi_{nN}\varphi_n   H   \varphi_{n'}\Phi_{n'N'})$
static approximation	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)})$	0	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)})$
Condon approximation	$v_{nn'}^{(0)} \frac{v_{n'n'}^{(0)} - v_{nn}^{(0)}}{U_n^{(0)} - U_{n'}^{(0)}} \times$ $\times (\Phi_{nN}^{(1)}   Q^2   \Phi_{n'N'}^{(1)})$	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)}) -$ $- v_{nn'}^{(0)} \frac{v_{n'n'}^{(0)} - v_{nn}^{(0)}}{U_n^{(0)} - U_{n'}^{(0)}} \times$ $\times (\Phi_{nN}^{(1)}   Q^2   \Phi_{n'N'}^{(1)})$	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)})$
non-Condon approximation	0	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)})$	$v_{nn'}^{(0)} (\Phi_{nN}^{(1)}   Q   \Phi_{n'N'}^{(1)})$

<sup>5</sup>) Strictly speaking, Kovarskii [9] used a somewhat different explicit form of the wave functions  $\varphi_n^{NC}$  in that one of the energies in the denominator in [22] is replaced by the exact value of a two-level model.

in the original paper by Kovarskii [9] can be easily avoided by means of the relations

$$\begin{aligned}
 B_{nNn'N'}^{\text{NC}} &= v_{nn'}^{(0)} \left( \Phi_{nN}^{(1)} \left[ H_{\text{L}}, \frac{Q}{\tilde{U}_{n'}(Q) - \tilde{U}_n(Q)} \right] \Phi_{n'N'}^{(1)} \right) + O(v_{nn'}^{(0)2}) = \\
 &= v_{nn'}^{(0)} \left( \Phi_{nN}^{(1)} (\tilde{U}_{n'}(Q) - \tilde{U}_n(Q)) \frac{Q}{\tilde{U}_{n'}(Q) - \tilde{U}_n(Q)} \Phi_{n'N'}^{(1)} \right) + O(v_{nn'}^{(0)2}) = \\
 &= v_{nn'}^{(0)} (\Phi_{nN}^{(1)} | Q | \Phi_{n'N'}^{(1)}) + O(v_{nn'}^{(0)2}), \quad (23)
 \end{aligned}$$

where, again, the equation of motion (15) and the conservation of the total energy were exploited. We remark that the more compact calculation of the transition amplitude within the non-Condon approach by Huang [27] and one of the present authors [28, 29] are based essentially on the same idea as (23). To summarise, apart from unimportant second-order corrections, we can represent our results in the following scheme (Table 1).

### 3. Conclusions

In this paper we have given the rigorous formulation of the adiabatic theory of multiphonon capture processes. The lack of former applications of the adiabatic theory is that the perturbation operator has been identified with the non-adiabatic part of the total Hamiltonian. This is only correct if exact adiabatic electron wave functions are used as the quasi-stationary states. In all practical calculations, however, approximate adiabatic solutions were taken. Then the perturbation operator includes in addition to the non-adiabatic part also contributions from the adiabatic part of the total Hamiltonian. In the case of Condon adiabatic wave functions this latter part yields even the main contribution to the transition matrix element. This contribution,  $A_{nNn'N'}$ , has been omitted in all foregoing papers relying on the Condon approximation, whereas the much smaller contribution,  $B_{nNn'N'}$ , due to the non-adiabatic part has been taken into account. As a consequence of this, the absolute value of the capture cross-section came out much too small. With the complete perturbation operator the same result is obtained in the adiabatic Condon approximation as in other approaches to first order in  $v_{nn'}^{(0)}$ . If the non-Condon functions according to [9, 30, 31] are taken as the stationary states the disregard of the adiabatic part within the perturbation operator remains still an incorrect step, but the relative weight of matrix elements  $A_{nNn'N'}$  and  $B_{nNn'N'}$  now is changed in favour of  $B_{nNn'N'}$  as compared to the Condon approximation and the correct result up to first order in  $v_{nn'}^{(0)}$  could be obtained by using exclusively the non-adiabatic part of the perturbation operator. The simplest way to calculate the capture cross-section up to first order in  $v_{nn'}^{(0)}$  is the static approximation. We have shown that the static result can be obtained as the lowest non-vanishing approximation within the adiabatic theory. With the static wave functions as the stationary states the non-adiabatic perturbation matrix element  $B_{nNn'N'}$  vanishes exactly and the transition rate is completely determined by the adiabatic term  $A_{nNn'N'}$ . Thus, former calculations based on the static approach can be considered to be correct also from the more general point of view of the present paper. Whether the static approach in the theory of multiphonon capture processes should be considered as an independent calculation scheme or as the lowest approximation of the adiabatic theory is to some extent a question of interpretation. As we have shown there is no need to leave the adiabatic theory to get the static result. On the other hand, in its general form the static approximation may be considered to provide an alternative decoupling procedure as compared with the adiabatic approximation

[21]. For practical calculations, however, only the lowest order of the static approximation has been used so far. To this order the static theory turns out to be a certain approximation within the adiabatic theory.

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(Received December 7, 1981)