Carrier-relaxation dynamics in intragap states: The case of the superconductor YBa₂Cu₃O_{7- δ **}** and the charge-density-wave semiconductor $K_{0.3}MoO₃$

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The unusual slow carrier relaxation dynamics—observed in femtosecond pump-probe experiments on hightemperature superconductors and recently also in a charge-density-wave system—is analyzed in terms of a model for relaxation of carriers in intragap states. The data on $YBa_2Cu_3O_{7-\delta}$ near optimum doping and $K_{0,3}$ MoO₃ are found to be described very well with the model using a BCS-like gap which closes at T_c . From the analysis of the data we conclude that a significant intragap density of localized states exists in these materials, which can be clearly distinguished from quasiparticle states by the time-resolved optical experiments because of the different time and temperature dependences of the photoinduced transmission or reflection. Localized charges are suggested to be the most likely origin of the intragap states, while the similarity of the response in the two materials appear to exclude spin and vortex excitations.

I. INTRODUCTION

Photoinduced absorption or reflection spectroscopy using femtosecond lasers is potentially a very powerful tool for the study of the electronic structure of superconductors and related materials. Recent pump-probe experiments on cuprate superconductors^{1–4} and some other materials⁵ have shown that a photoinduced change in absorption or reflection can be observed at low temperatures and especially for $T < T_c$. The effects are believed to be caused by excited-state absorption of the probe pulse from photoexcited quasiparticle (QP) states⁴ and theoretical analysis of the response was found to be in good agreement with experimental data on $YBa₂Cu₃O_{7-\delta}$ over a wide range of doping.⁴ However, in addition to the QP response which occurs on the picosecond and subpicosecond time scale, a distinct slower response was also consistently observed in $YBa_2Cu_3O_{7-\delta}$ (YBCO),² $Bi_2Sr_2CaCu_2O_8$ (BISCO), and $Bi_2Y_xCa_{1-x}SrCu_2O_8$, and more recently in the charge-density-wave (CDW) quasi-onedimensional Peierls insulator $K_{0,3}$ MoO₃.⁵ It was thought to be of nonthermal origin (detailed analysis is given in Ref. 6) and occurs on a time scale of 10^{-8} s or longer (see Fig. 1 for details). Its anomalous *T* dependence² — which is qualitatively different from the *T* dependence of the fast QP recombination dynamics — led the authors to the suggestion that the signal is due to localized states near the Fermi energy. However, the processes involved were so far not discussed in any detail.

In this paper we examine quantitatively the photoinduced absorption (or reflection) from localized intragap states. We develop a theoretical model for two different cases: (i) the case of a material with a BCS-like collective gap, and (ii) the case of a *T*-independent gap (sometimes called a pseudogap) which exists above T_c . We compare the model predictions with the available data on the temperature dependence of the photoinduced relaxation on the nanosecond time scale in two different materials, both of which are generally thought to have a low-temperature gap: $YBa₂Cu₃O_{6.9}$ ($T_c=90$ K) and $K_{0.3}$ MoO₃ (T_c =183 K).

II. THEORETICAL MODEL

As the name implies, pump-probe spectroscopy involves the excitation of the material by an ultrashort pump laser pulse and the subsequent measurement of the resulting *change* in optical absorption, transmission or reflection of the sample caused by photoexcited charge carriers. As photons from the pump laser pulse are absorbed, they excite electrons and holes in the material see schematic diagram in Fig. $2(a)$. These particles release their extra kinetic energy by scattering amongst themselves and with phonons [step 2 in Fig. $2(a)$]. This energy relaxation process is very rapid and the particles end up in QP states near the Fermi energy (E_F) within $\tau_e = 10 - 100$ fs.⁴ Subsequent relaxation is slowed down by the presence of the gap and a relaxation bottleneck

FIG. 1. A photoinduced transmission signal $\Delta T/T$ as a function of time *t* after photoexcitation in $YBa_2Cu_3O_{7-\delta}(T_c=90 \text{ K})$ taken at $T=80$ K (points), together with the fit (solid line). (1) is the baseline signal with no pump applied. (2) is the long-lived signal pileup remaining from previous pulses, (3) (dashed line) is the signal due to QP recombination, and (4) (dotted line) is the long-lived signal remaining after all the QP signal has decayed. Signal pileup contribution and the single pulse contribution to the slow photoinduced signal are given by A and a_0 , respectively.

is formed. From pump-probe photoinduced transmission experiments in YBCO (Refs. 1 and 4) and BISCO, $⁶$ the relax-</sup> ation times of the photoexcited QP's were found to be in the range $\tau_{OP} = 0.3-3$ ps.

In addition to the picosecond transient, the signal on nanosecond time scale has been consistently observed in $HTSC₁^{2,6,11}$ and recently also in quasi-one-dimensional CDW insulator $K_{0.3}MoO₃$.⁵ In Fig. 1 the photoinduced transient taken on YBCO at $T=80$ K is shown (squares). After photoexcitation $(t=0)$ the signal relaxes within 10 ps to some nonzero value, that can be represented by the constant on the 100 ps time scale.⁶ The lifetime of the slow component τ_L cannot be directly measured, since it appears to be longer than the interpulse separation of $t_r \approx 10-12$ ns. This results in the signal pileup due to accumulation of the response from many pulses. The magnitude of this pileup, *A*, given by the difference in the signal amplitude when the pump pulse is unblocked (2) , and zero signal, when the pump is blocked (1) at negative time delays (see Fig. 1), is several times larger than the single pulse contribution, a_0 , given to be the difference between (4) and (2) .

Assuming — for simplicity — that relaxation of the signal is exponential, we can write an equation for the steadystate amplitude *A*:

$$
A = a_0 \sum_{n=0}^{\infty} \exp(-nt_r/\tau_L) = a_0/[1 - \exp(-t_r/\tau_L)]. \quad (1)
$$

Usually, $A \ge a_0$ and we can expand the exponent in the denominator of Eq. (1), to obtain $\tau_L = t_r(A/a_0)$. From the experiments^{2,5} it appears that $\tau_l > 10^{-7}$ s. This is long in comparison with the phonon relaxation time and with the phonon escape time from the excitation volume into the bulk or thin-film substrate, which is typically 10^{-10} s, so we can ignore phonon escape effects and discuss only intrinsic relaxation processes.

The process giving rise to the actual photoinduced optical signal from intragap states is shown as step 3 in Fig. 2(a). Here we do not discuss the optical probe process in detail, but make the very general assumption that the photoinduced *change* in sample transmission $\Delta T/T$ or reflection $\Delta R/R$ is proportional to the photoinduced density of localized states populated by the laser pump pulse. The photoinduced signal $\Delta T/T$ (or $\Delta R/R$) is then proportional to the number of filled

FIG. 2. (a) The pump-probe optical diagram. *1* represents the pump pulse exciting charge carriers into a higher-lying band, *2* the carriers rapidly relax their energy to states near the Fermi energy. *3* represents the probe pulse. (b) A schematic diagram of the terms contributing to the relaxation of intragap states in a superconductor with a gap 2Δ . In the case of a CDW gap, the $e-h$ pairs take the place of Cooper pairs, so the rate equation remains the same.

localized states and the *T* dependence is mainly determined by the occupation of the intragap states.

Since the relaxation time τ_L is long in comparison with τ_{OP} and phonon relaxation times, we assume that phonons and quasiparticles can be described by equilibrium densities N_{ω} and *N*, respectively. For the relaxation of the localized carriers we apply arguments similar to those originally proposed by Rothwarf and Taylor.⁷ The rate equation for the total density of localized excitations N_L is then given by

$$
\frac{dN_L}{dt} = -RN_L^2 - \tilde{\gamma}N_L + \gamma N + \beta N_\omega.
$$
 (2)

The first term in Eq. (2) describes the recombination of two localized excitations to a Cooper pair with a recombination rate *R*. The second and third terms describe the exchange of an electron or a hole between the localized and quasiparticle states with density N_L and N , respectively, and with a rates $\tilde{\gamma}/\gamma \propto \exp(-\Delta E/k_BT)$ where ΔE is the energy barrier between trapped carriers and quasiparticles⁸. The last term describes the spontaneous creation of localized excitations by phonons with a relaxation rate β . The four processes in Eq. (2) are shown schematically in Fig. $2(b)$.

Assuming the ansatz for $N_L = N_{L0} + n_L$, where N_{L0} is equilibrium density of localized particles and n_L is the photoinduced density created by the laser pulse and taking into account that N_{ω} and *N* are given by their equilibrium values, we can rewrite Eq. (2) :

$$
\frac{dn_L}{dt} = -Rn_L^2 - (2RN_{L0} + \tilde{\gamma})n_L.
$$
 (3)

This equation is sufficiently general that it can be applied to different superconductors and different gaps. (For the case of a CDW gap, the Cooper pairs are replaced by $e-h$ pairs.) In this paper we consider (a) a superconductor with a BCS-like gap (which can also be used in the case of a gap formed by a CDW) and (b) a Bose condensate of preformed pairs with a *T* independent gap. In the latter case, the gap is better considered as an energy-level splitting between paired and unpaired states, and the main difference is that the gap is *T* independent and exists above T_c . A general exact solution to Eq. (3) is given in the Appendix. Next we consider the limiting behavior at low T and near T_c .

In the case of a collective BCS-like gap $\Delta_s(T)$, the recombination rate below T_c is to the lowest order in Δ_s proportional to the square of the order parameter: $9,10$

$$
R \simeq \alpha [\Delta_s(T)/\Omega_c]^2, \tag{4}
$$

where α is a constant, and Ω_c is the phonon spectrum cutoff frequency. [Above T_c all recombination processes disappear as $\Delta_{s}(T) \rightarrow 0$.] To relax the carriers in localized intragap states via quasiparticle states [the term $\tilde{\gamma}n_L$ in Eq. (3)], an energy of the order of Δ _s is required and so this process is exponentially suppressed at low temperatures. The term proportional to N_{L0} is also small at low temperatures, because the number of thermally excited localized excitations is small as $\left[k_B T/\Delta_s(0)\right]^{\mu}$, where μ depends on the density of localized states. Therefore, for analysis of the relaxation of localized excitations at low temperatures $T \ll T_c$, we retain only the first term in Eq. (3) giving a solution of the form

$$
n_L(t) = n_L(0) / [n_L(0)Rt + 1].
$$
 (5)

To obtain the stationary solution for a repetitive laser pump pulse train excitation, we use the condition that the total number of localized excitations that recombine between two laser pulses should equal the number of localized excitations created by each laser pulse:

$$
n_L(0) - n_L(0) / [n_L(0)Rt_r + 1] = \eta n_{ph}(T), \tag{6}
$$

where $\eta \propto \gamma \tau_{OP} = \eta'/\Delta(T)$ is the probability of trapping a QP into a localized state and $n_{OP}(T)$ is the number of photoinduced QP's at temperature *T* created by each laser pulse. Since the number of photoexcited carriers is typically small compared to the overall carrier density $\eta n_{OP} \le n_L(0)$, we can estimate $n_L(0)$ as

$$
n_L(0) = \sqrt{\frac{\eta n_{QP}(T)}{Rt_r}}.\t(7)
$$

As a result, combining Eqs. (4) and (7) we get an expression for the *T* dependence of the photoinduced transmission (or reflection) amplitude *at low temperatures*:

$$
|\Delta T/T| \propto n_L(0) = \sqrt{\frac{\eta n_{QP}(T)}{\alpha t_r}} \frac{\Omega_c}{\Delta_s(T)},
$$
\n(8)

where n_{OP} for a BCS-like case is given by⁴

 $n_{OP}(T)$

$$
= \frac{\mathcal{E}_I/(\Delta_s(T) + k_B T/2)}{1 + [2 \nu/N(0) \hbar \Omega_c] \sqrt{2k_B T/\pi \Delta(T)} \exp[-\Delta_s(T)/k_B T]}.
$$
\n(9)

Here \mathcal{E}_I is the energy density deposited per pulse, ν is the effective number of phonons per unit cell involved in the relaxation process and $N(0)$ is the density of states at the Fermi energy in units eV^{-1} cell⁻¹ spin⁻¹.

Near T_c , when the number of thermally excited localized carriers becomes comparable or larger than number of nonequilibrium carriers, the relaxation terms $2RN_{L0}n_L$ and $\tilde{\gamma}n_L$ become dominant, and the solution to Eq. (3) has an exponential rather than a power law time dependence of the form

 $n_L(t) = n_L(0) \exp(-t/\tau)$, where $1/\tau = [2R(T)N_{L0} + \tilde{\gamma}]$. For $T \rightarrow T_c$, the temperature dependence of the photoinduced signal amplitude is then given by

$$
|\Delta T/T| \propto n_L(0) = \frac{\eta n_{QP}(T)}{[2N_{L0}R(T) + \tilde{\gamma}]t_r}.
$$
 (10)

Thus the predicted amplitude of the signal increases with increasing *T* up to T_c and drops to zero above T_c .

In the case of a *T*-independent gap (or "pseudogap") Δ^p , we assume that the gap exists at all temperatures. Since the gap does not close at T_c , the recombination rate does not go to zero at T_c and instead of Eq. (4) we have a constant Γ .

However, below T_c the presence of the condensate may also have an effect on the recombination of localized excitations. In general, the relaxation rate is a function of the order parameter. To take this into account, we can expand it in terms of even powers of Δ . Near T_c the order parameter is small and we can keep only the lowest power in Δ^{2} .¹³ If we assume that the order parameter exhibits mean-field behavior $(\Delta \propto \sqrt{1-T/T_c})$, then

$$
R \simeq \widetilde{\alpha}(1 - T/T_c) + \Gamma,\tag{11}
$$

where $\tilde{\alpha}$ is phenomenological constant which describes the dependence of the relaxation rate on T below T_c , which in general is not equal to 0.

A slightly different expression for *R* is obtained if we assume that the recombination rate is dependent on the pair momentum in the condensate through the kinetic energy. To illustrate this, we write the recombination rate as $\Gamma = \Gamma_0$ $+ \overline{\Gamma}$ where $\overline{\Gamma}$ is the momentum averaged recombination rate, and Γ_0 is the recombination rate for pairs with $k=0$. The recombination rate is generally proportional to the number of pairs in the condensate n_p . For Bose condensation this is given by $n_p \propto [1 - (T/T_c)^{3/2}]$, and we thus obtain a formula for the total relaxation rate which is similar to Eq. (11) , but with a different temperature dependence in the first term:

$$
R = (\Gamma_0 - \bar{\Gamma}) [1 - (T/T_c)^{3/2}] + \bar{\Gamma}.
$$
 (12)

In principle the two cases (11) or (12) can be distinguished by measurements of the *T* dependence of the photoinduced transmission or reflection below T_c , although the difference will be very small and very high quality data is needed to do this.

To obtain the photoinduced signal amplitude, we substitute Eq. (11) into Eq. (3) :

$$
|\Delta T/T| \propto n_L(0) = \sqrt{\frac{\nu n'_{QP}}{\{\tilde{\alpha} [1 - (T/T_c)^{\beta}] + \Gamma\} t_r}},\qquad(13)
$$

where β is a constant (generally 1 or 3/2) and n'_{QP} is the equivalent of formula (9) for the case of a temperatureindependent gap:⁴

$$
n'_{QP} = \frac{E_I/\Delta^p}{1 + [2\nu/N(0)\hbar\Omega_c] \exp(-\Delta^p/k_B T)}.
$$
 (14)

In contrast to the BCS case, expression (13) is nonzero above T_c and reduces to

$$
|\Delta T/T| \propto n_L(0) = \sqrt{\frac{\nu n'_{QP}}{\Gamma t_r}}, \qquad (15)
$$

which implies that the photoinduced absorption signal should remain observable well above T_c and should reveal the presence of a pseudogap if it exists.

Just as before, a crossover to exponential time relaxation takes place when the number of thermally excited excitations become large and the second term in Eq. (3) becomes dominant, i.e., $T \sim \Delta^p$, leading to a linear intensity dependence of $|\Delta T/T|$ given by modified Eq. (10) with n'_{OP} and *R* from Eq. (12) . Note that this crossover may occur at higher temperatures than experimentally measured since *T*-independent gap is typically $\Delta^p \ge 300$ K.

III. COMPARISON WITH EXPERIMENTAL DATA

The temperature dependence of $|\Delta T/T|$ or $|\Delta R/R|$ for optimally doped YBa₂Cu₃O_{7- δ} (Refs. 2 and 11) and for $K_{0,3}$ MoO₃ (Ref. 5) is plotted in Figs. 3(a) and 3(b), respectively. In both compounds the signal amplitude increases with increasing temperature followed by an abrupt drop above T_c . Now we compare the predicted *T* dependences of $|\Delta T/T|$ [Eqs. (8) and (10)] with the data. The parameters used in the fits are the same as previously used in the analysis of the fast relaxation component^{4,5} with values of dimensionless constant $2\nu/N(0)\hbar\Omega_c \approx 30$ for YBa₂Cu₃O_{7- δ} (Ref. 4) and \simeq 10 for $K_{0.3}$ MoO₃.⁵ Since the magnitude of the gap is of the order of $\Delta_c(0) \sim 5kT_c$ we expect that Eq. (8) is valid up to temperatures close to T_c . In Figs. 3(a) and 3(b) we show the calculated temperature dependences of $|\Delta T/T|$ using Eq. (8) in comparison with experimental data for $YBa₂Cu₃O_{7-\delta}$ and $K_{0.3}MoO₃$, respectively. We would like to stress that Eq. (8) is independent of the shape of density of states (DOS) of localized states within the gap and shows a universal temperature dependence. It can be seen from Fig. $3(b)$ that at low temperature there is deviation of the calculated curve from the experimental points. To explain this effect we should remember that Eq. (4) for the rate *R* is valid near T_c where $\Delta_c(T)$ is small. [If we add next — fourthorder — term in the expansion of *R* in powers of $\Delta_c(T)$ we can account for this discrepancy.

Near T_c Eq. (8) fails and Eq. (10) should be used (see, also, the Appendix). It leads to a crossover from square-root intensity dependence of $|\Delta T/T|$ at low *T* described by Eq. (8) to linear intensity dependence near T_c predicted by Eq. $(10).$

Finally let us discuss the effect of $\tilde{\gamma}$. In Figs. 3(a) and 3(b) fits to the data using the general solution $[Eq. (A3)]$ are shown with dashed lines. In these fits we have also added the fourth-order term in the expansion of *R* in powers of $\Delta_c(T)$. As can be seen from these fits the effect of $\tilde{\gamma}$ becomes important near T_c by cutting the divergence of $|\Delta T/T|$ as *T* $\rightarrow T_c$.

In Fig. 3(c) we have plotted calculated values of $\left|\Delta T/T\right|$ ${}^{\alpha}n_L(0)$ for the case of temperature-independent pseudogap (which might be applicable in underdoped cuprates, for example) as a function of temperature for different values of parameter α using Eq. (13). As can be seen from Fig. 3(c), in this case slow relaxation via localized states is present also

FIG. 3. (a) The temperature dependence of the photoinduced absorption from localized states in $YBa₂Cu₃O_{6.9}$ taken from Ref. 2 (open circles) and Ref. 11 (solid circles). The solid line is a plot of expression (8) with $\Delta_c(0)/k_BT_c=5$ whereas the dashed line represents a general solution [Eq. (A3)] with nonzero $\tilde{\gamma}$ term and additional fourth-order term in the expansion *R* in powers of $\Delta_c(T)$. (b) The temperature dependence of the photoinduced reflection in $K_{0,3}MoO₃$ from Ref. 5 (open circles) compared with the model fit using Eq. (8) with $\Delta_c(0)/k_B T_c$ =4.8 (solid line). The general solution $[Eq. (A3)]$ is represented by the dashed line. (c) The calculated temperature dependence of the photoinduced absorption from localized states in case of *T*-independent pseudogap Eq. (13) with $\Delta^p/k_BT_c=8$ and different α/Γ ratios.

above T_c . This effect is due to temperature dependence of n'_{OP} controlled by *T*-independent pseudogap above T_c .

In cuprate superconductors there is spectroscopic evidence suggesting that there is a significant density of states in the gap possibly extending to the Fermi level, which is often attributed to a *d*-wave gap symmetry. However, by normal spectroscopies it is difficult to determine if the states in the gap are QP states or, for example, localized states.

Time-resolved spectroscopy can answer this question rather effectively because of the different time- and temperaturedependences of the QP's and localized carrier relaxations. It was argued that in the presence of impurity scattering QP DOS in the *d*-wave state remains finite at zero energy.¹⁴ Recently it was proposed¹⁵ that the quasiparticles in the superconducting state may become strongly localized for short coherence length *d*-wave superconductors. However, this statement has been questioned by Balatsky and Salkola¹⁶ and remains controversial. On the basis of available experimental data we cannot make any definite conclusion about *origin* of intragap localized states.

We can, however, estimate the density of the intragap states from the available data by assuming that the optical probe process (step 3) is similar for excited state absorption from localized states and for QP's. Both optical probe processes involve allowed transitions to the same final state E_2 and so this assumption is not unreasonable. From typical photoinduced reflection data for YBCO (as in Fig. 1), we find that approximately $|\Delta T/T|_{L} \approx |\Delta T/T|_{OP}$, implying that also $n_l(0) \approx n_{OP}$. From this we can conclude that the density of intragap states is *comparable* with the density of QP states. This observation has important implications for the interpretation of frequency-domain spectroscopies, since it suggests that the spectra should show a very significant intragap spectral density due to localized states, irrespective of the gap symmetry.

Assuming that the optical transition probability of the probe pulse is the same for QPs as for the intragap states, from Eqs. (8) and (9) we obtain

$$
\frac{|\Delta T/T|_L}{|\Delta T/T|_{qp}} \sim \frac{n_L}{n_{QP}} = \frac{\eta \tau_L}{t_r},\tag{16}
$$

where $|\Delta T/T|_{qp}$ is the photoinduced transmission due to the QP's. From Fig. 1 typically $n_L/n_{QP} = 0.1-1$ and using a pulse repetition rate $t_r = 12$ ns and with $\tau_l = 100$ ns, we obtain an estimate of the trapping probability for carriers by localized states of $\eta=0.1-1$. This estimate should be considered as an upper limit of η since we do not take into account the differences in dipole matrix elements for the transition between localized and QP states.

A detailed discussion of the origin of the localized intragap states in the cuprates should be deferred until more systematic data as a function of doping is available, and we only mention some of the most likely possibilities: (i) localized states associated with the inhomogeneous ground state of the cuprates (stripes),¹⁷ (ii) intrinsic defect states, (iii) localized QP states in d -wave superconductor,¹⁵ and, possibly, (iv) holons.¹⁸ In $K_{0.3}MoO₃$, the nature of intragap excitations has been a subject of extensive study over the years and the reader is referred to Ref. 12 for a review. However, the fact that the signals in $K_{0,3}MoO₃$ and $YBa₂Cu₃O_{7-\delta}$ are very similar appears to rule out both spin excitations and vortex states, leaving localized charges as the most plausible origin of the intragap states.

IV. CONCLUSIONS

To conclude, the calculated time and temperature dependence of the photoinduced absorption for the case of a BCSlike gap is found to be in good agreement with experimental data from femtosecond time-resolved spectroscopy on the cuprate superconductor $YBa₂Cu₃O_{7-\delta}$ near optimum doping and the charge-density-wave insulator $K_{0,3}MoO₃$. We find that time-resolved spectroscopy can very effectively distinguish between QP states and localized states in the gap. A rather surprising feature of the data is the remarkable separation of the QP response on the femtosecond time scale and the slow response of intragap state relaxation on the scale of hundreds of nanoseconds. In both materials we find a significant intragap density of states, which displays very different time dynamics and *T* dependence than the QP states above the gap.

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APPENDIX

Analytic solution of Eq. (3) has the form

$$
n_L(t) = \frac{2N_{L0}C \exp(-t/\tau)}{1 - C \exp(-t/\tau)}.
$$
 (A1)

Here $1/\tau = 2N_{L0}R + \tilde{\gamma}$. Constant *C* can be found from the following equation [see also Eq. (6)]:

$$
n_L(0) - n_L(t_r) = \eta n_{QP}.
$$
 (A2)

Combining these two equations one obtains the following form for $n_L(0)$:

$$
n_L(0) = N_{L0}(1 + \tilde{\gamma}/2RN_{L0})
$$

$$
\times \left[\sqrt{1 + \frac{\eta n_{QP}}{N_{L0}^2 (1 + \tilde{\gamma}/2RN_{L0})^2 R t_r}} - 1 \right].
$$
 (A3)

This solution reduces to Eq. (8) if $\eta n_{QP} / N_{L0}^2 (1)$ $+\tilde{\gamma}/2RN_{L0})^2Rt_r\geq 1$ and to Eq. (10) in the opposite limit.

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