Tomonaga's Model and the Threshold Singularity of X-Ray Spectra of Metals

K. D. SCHOTTE* AND U. SCHOTTE†
University of California, San Diego, La Jolla, California 92037
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Singularities near the threshold of the soft x-ray spectra of metals have been predicted by Mahan and have recently been calculated by Nozières *et al.* using the model of a localized core hole. We show that the singular behavior can be understood in terms of density waves of the conduction electrons which are excited when, in the absorption process, the core hole is created, providing an attractive potential for the conduction electrons. For the description of the conduction electrons in terms of density waves, Tomonaga's model is adopted.

1. INTRODUCTION

IN the x-ray absorption process, a deep-lying core electron is excited into the conduction band by an incoming x-ray photon. The core hole left behind acts as a one-body potential on the conduction electrons. A simplified model has been used to describe the situation[1]:

$$H=\sum_k \epsilon_k a_k^\dagger a_k+E_0 b^\dagger b+\frac{1}{N}\sum_{k,k'} V_{kk'} a_k^\dagger a_{k'} b b^\dagger. \quad (1)$$

Here the hole (described by $b^\dagger$,b) is represented by a single nondegenerate level E_0 with infinite lifetime. It is assumed to interact with the free conduction electrons (described by $a_k^\dagger$, a_k) via a contact potential $V_{kk'}=V$; the interaction between conduction electrons has been neglected.

We confine ourselves to the discussion of the adsorption process. In this case the transition role according to the Golden Rule is

$$W(\omega)=2\pi\sum_f\left|\sum_k w_k(\omega)\langle f|a_k^\dagger b|i\rangle\right|^2\delta(\tilde{E}_i+\omega-\tilde{E}_f). \quad (2)$$

For simplicity the matrix elements of the dipole operator $w_k(\omega)$ will be regarded as constant. In an equivalent one-body description, the transition rate is given by

$$W(\omega)=2\pi w^2\sum_f\left|\left\langle f_{n+1}\left|\frac{1}{\sqrt{N}}\sum_k a_k^\dagger\right|i_n\right\rangle\right|^2 \times\delta(E_i-E_f+E_0+\omega). \quad (3)$$

The initial state

$$|i_n\rangle=\prod_{k=0}^{k_F} a_k^\dagger|0\rangle$$

is the n-particle ground state of the Hamiltonian

$$H_i=\sum_k \epsilon_k a_k^\dagger a_k, \quad (4)$$

with the energy $E_i=\tilde{E}_i-E_0$. The final states $|f_{n+1}\rangle$ are the $(n+1)$-particles eigenstates of the Hamiltonian

$$H_f=\sum_k \epsilon_k a_k^\dagger a_k+\frac{V}{N}\sum_{k,k'} a_k^\dagger a_{k'}. \quad (5)$$

The final states $|f_{n+1}\rangle$ hold the clue to the problem. According to Anderson,[2] the overlap between the initial and any final state containing a finite number of electron-hole pairs is zero in the limit of infinite volume.

So one has the choice of doing the calculations with finite volume, performing the limiting process to infinite volume in the end, or of circumventing the problem to determine the final states by using Green's-function techniques.[1] However, the physical origin of the singular behavior of the transition rate near the threshold[1,3] does not seem to be clearly understood.

In this work, we offer another way of calculating the response of the free electrons to the sudden switching on of the core hole potential. We will use the Tomonaga model[4] by which we can describe the excitations of the Fermi sea in terms of density waves. On the one hand, we circumvent the difficulty posed by the Anderson theorem, and, on the other hand, we have a plausible physical interpretation of what is going on.

2. FORMULATION OF THE PROBLEM IN TERMS OF TOMONAGA'S BOSONS

We introduce the density operator

$$\rho_k=\sum_{k_1=0}^{k_D-k}\frac{1}{\sqrt{N}}a_{k_1}^\dagger a_{k_1+k}, \qquad \rho_{-k}=\sum_{k_1=k}^{k_D}\frac{1}{\sqrt{N}}a_{k_1}^\dagger a_{k_1-k}, \quad k\gtrless 0 \quad (6)$$

and consider a band of width D, assuming a constant density of states within the band. $k=0$ and $k_D=k$ are the momenta at the bottom and at the top of band, respectively. The electron energies are given by

$$\epsilon_k=(k-k_F)/\rho, \quad (7)$$

where ρ is the density of states.

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[1] P. Nozières and C. T. de Dominicis, Phys. Rev. **178**, 1084 (1969); B. Roulet, I. Gavoret, and P. Nozières, *ibid.* **178**, 1252 (1969).

[2] P. W. Anderson, Phys. Rev. Letters **18**, 1049 (1967).

[3] G. D. Mahan, Phys. Rev. **163**, 612 (1967).

[4] S. Tomonaga, Progr. Theoret. Phys. (Kyoto) **5**, 544 (1950).

Actually, in the Tomonaga model the free-particle energy has to be proportional to the momentum. Another important feature of the Tomonaga model is its one-dimensionality. But the problem considered here can be looked upon as a one-dimensional one because it involves only s-wave scattering. So the summation in (6) over k_1 is to be understood as a summation over energy shells. We examine now the commutation relations of the ρ_k's:

$$[\rho_k,\rho_{k'}]=0 \quad \text{for} \quad k,k'>0 \quad \text{and} \quad k,k'<0, \tag{8}$$

$$[\rho_k,\rho_{-k'}]=\frac{1}{N}\sum_{k_1=0}^{k'} a_{k_1}{}^{\dagger}a_{k_1+k-k'}-\frac{1}{N}\sum_{k_1=k_D-k}^{k_D-k+k'} a_{k_1}{}^{\dagger}a_{k_1+k-k'}$$

$$\text{for} \quad k>k'>0, \tag{9}$$

$$[\rho_k,\rho_{-k}]=\frac{1}{N}\sum_{k_1=0}^{k} a_{k_1}{}^{\dagger}a_{k_1}-\frac{1}{N}\sum_{k_1=k_D-k}^{k_D} a_{k_1}{}^{\dagger}a_{k_1}. \tag{10}$$

We adopt Tomonaga's approximation and substitute these complicated commutation relations by simpler ones:

$$[\rho_k,\rho_{-k'}]=k\delta_{kk'}. \tag{11}$$

For a detailed discussion of this important step we refer to Tomonaga's paper. In short, the simpler commutation relations may be used if only a certain subspace of all possible states has to be considered. These are the states which do not have unoccupied levels deep in the bottom of the band or occupied levels high above the Fermi energy at the top of the band, such that the parts left out of the commutator do not contribute very much. In addition, we have to assume that V be very small compared to the bandwidth. On the one hand we had to consider a contact potential to keep mathematics simple, but, on the other hand, a contact potential causes excitations deep below and high above the Fermi level (or short density waves), which is explicitly excluded in the model. So we must expect all our results to be valid only for small V.

With (11) we calculate the commutator

$$[H_i,\rho_k]=-(k/\rho)\rho_k, \tag{12}$$

where use has been made of the linear energy-momentum dependence, and we conclude that H_i has the form[5]

$$\tilde{H}_i=\sum_{k>0}\frac{1}{\rho}\rho_{-k}\rho_k. \tag{13}$$

The transcription of H_f [Eq. (5)] in terms of bosons also leads to a very simple expression,

$$\tilde{H}_f=\sum_{k>0}\frac{1}{\rho}\rho_{-k}\rho_k+\frac{V}{\sqrt{N}}\sum_{k>0}(\rho_k+\rho_{-k}). \tag{14}$$

[5] The tacit assumption is the completeness of the k, which is only fulfilled in the subspace of s-wave states.

It is convenient to introduce the normalized boson operators ($k>0$)

$$b_k=(1/\sqrt{k})\rho_k, \quad b_k{}^{\dagger}=(1/\sqrt{k})\rho_{-k}, \tag{15}$$

which obey the usual boson commutation relations

$$[b_k,b_{k'}{}^{\dagger}]=\delta_{kk'}. \tag{16}$$

In terms of bosons

$$\tilde{H}_i=\sum_{k>0}\frac{k}{\rho}b_k{}^{\dagger}b_k, \tag{17}$$

and

$$H_f=\sum_{k>0}\frac{k}{\rho}\left(b_k{}^{\dagger}+\frac{V}{\sqrt{(kN)}}\right)\left(b_k+\frac{V}{\sqrt{(kN)}}\right)-\frac{V^2\rho}{N}\sum_k 1. \tag{18}$$

$\tilde{H}_i$ and $\tilde{H}_f$ describe a set of harmonic oscillators. The effect of the potential is a shift of the zero points of the harmonic oscillators. As the transformation procedure is only correct up to a constant we will drop the constant term on the right-hand side of Eq. (18) in the following: In order to calculate the transition rate (3) we have to express the operator

$$a^{\dagger}=\frac{1}{\sqrt{N}}\sum_{k_1}a_{k_1}{}^{\dagger}$$

in terms of harmonic-oscillator coordinates. This is a rather delicate problem. First we examine the commutation relations between $a^{\dagger}$ and the density operators ρ_k. We find

$$[\rho_k,a^{\dagger}]=\frac{1}{\sqrt{N}}a^{\dagger}-\frac{1}{N}\sum_{k_1=k_D-k}^{k_D}a_{k_1}{}^{\dagger} \quad \text{for} \quad k>0, \tag{19}$$

$$[\rho_{-k},a^{\dagger}]=\frac{1}{\sqrt{N}}a^{\dagger}-\frac{1}{N}\sum_{k_1=0}^{k}a_{k_1}{}^{\dagger} \quad \text{for} \quad k>0. \tag{20}$$

We simplify these commutation relations in the spirit of Tomonaga's approximation, i.e., the matrix elements

$$\left\langle f_{n+1}\left|\frac{1}{N}\sum_{k_1=0}^{k}a_{k_1}{}^{\dagger}\right|i_n\right\rangle$$

are considered to be small. This is correct within the Tomonaga model and means that the energy of the final states has to be small compared to the bandwidth. So, we shall use

$$[\rho_k,a^{\dagger}]=(1/\sqrt{N})a^{\dagger} \quad \text{for all } k>0 \tag{21}$$

instead of the exact commutation relations [Eqs. (19) and (20)]. Defining

$$U=\exp[\sum_k \alpha_k(b_k{}^{\dagger}-b_k)], \quad \alpha_k \text{ real}, \tag{22}$$

we have a canonical transformation which acts as a translation operator on any function of $b_k{}^{\dagger}$ and b_k,

namely,

$$U^\dagger b_k U = b_k + \alpha_k \quad \text{or} \quad [b_k, U] = \alpha_k U. \tag{23}$$

This relation is identical to (21), if we put $\alpha_k = 1/\sqrt{(kN)}$ and

$$a^\dagger \propto \exp\left[\sum_k \frac{1}{\sqrt{(kN)}}(b_k^\dagger - b_k)\right]. \tag{24}$$

3. TRANSITION PROBABILITY IN THE DENSITY WAVE MODEL

Before calculating the transition rate, let us ask how to interpret the Golden Rule [Eq. (3)] in the new description. The initial state is the lowest eigenstate of a set of harmonic oscillators [Eq. (17)]—the analog to the "quiescent" Fermi sea. The final states are all possible excitations of another set of oscillators with shifted zero points [see Eq. (18)]. The ground state of this second set is also the analog to a quiescent Fermi sea, which is (in the language of the fermion description) a Slater determinant of scattering waves. Excitations in both oscillator systems can be pictured as density waves.

According to (24), $a^\dagger$ acts as a shift operator distorting, say, the initial quiescent Fermi sea. That is, by injecting the core-state electron into the band, a local inhomogeneity of the electron density is created, by which each oscillator is elongated by a small amount. At the same time, the oscillators have to react to the core-hole potential V produced in the absorption process. This is described by the zero-point shift of the second set of oscillators. As will be seen later these two effects are additive. This picture of the problem has some similarity to the "small polaron," where now the real lattice is replaced by the band electrons, and the real phonons are replaced by Tomonaga's "phonons."

Instead of the transition rate [Eq. (3)], we calculate the correlation function

$$\mathfrak{F}(t) = \langle i | \exp(+i\tilde{H}_i t)\ a \exp(-i\tilde{H}_f t)\ a^\dagger | i \rangle, \tag{25}$$

which is related to $W(\omega)$ by

$$W(\omega) \propto \operatorname{Im} \frac{i}{\pi} \int_0^\infty e^{i(\omega + E_0)t} \mathfrak{F}(t) dt. \tag{26}$$

The canonical transformation

$$U_V = \exp\left[V\rho \sum_k \frac{1}{\sqrt{(kN)}}(b_k^\dagger - b_k)\right] \tag{27}$$

transforms $\tilde{H}_i$ into $\tilde{H}_f$:

$$\tilde{H}_f = U_V^\dagger \tilde{H}_i U_V, \tag{28}$$

so that

$$\mathfrak{F}(t) = \langle i | \exp(+i\tilde{H}_i t) a U_V^\dagger \exp(-i\tilde{H}_i t) U_V a^\dagger | i \rangle, \tag{29}$$

with

$$U a^\dagger \equiv B = \exp\left[(1+V\rho) \sum_k \frac{1}{\sqrt{(kN)}}(b_k^\dagger - b_k)\right] \tag{30}$$

and

$$B(t) = \exp\left[(1+V\rho) \sum_k \frac{1}{\sqrt{(kN)}} \times (b_k^\dagger e^{+i(k/\rho)t} - b_k e^{-i(k/\rho)t})\right]. \tag{31}$$

We have

$$\mathfrak{F}(t) = \langle i | B^\dagger(t) B(0) | i \rangle, \tag{32}$$

which is equal to[6]

$$\mathfrak{F}(t) = \exp\left((1+V\rho)^2 \frac{1}{N} \sum_{k>0} \frac{1}{k}(e^{-i(k/\rho)t} - 1)\right); \tag{33}$$

converting the sum in an integral, introducing a cutoff[7]

$$\frac{1}{N} \sum_{k>0}^{k_{\max}} \frac{1}{k}(e^{-i(k/\rho)t} - 1) = \int_0^{it k_{\max}(1/\rho)} \frac{e^{-x} - 1}{x} dx, \tag{34}$$

we find as the leading term for large t

$$\mathfrak{F}(t) \sim [t^{(1+V\rho)^2}]^{-1}. \tag{35}$$

Inserting $-V\rho = \delta_B/\pi$, where δ_B is the phase shift in Born approximation, we finally get for the energy dependence near the threshold

$$W(\omega) \sim (\omega + E_0)^{-[2\delta_B/\pi - (\delta_B/\pi)^2]}. \tag{36}$$

Thus, we have recovered the main feature of Nozière's result, namely, the singular threshold behavior of the response function. But in the correct answer δ_B is replaced by δ, the exact phase shift. In this context it is amusing to note that in the Luttinger model,[8] where an infinite energy spectrum with a linear dispersion is assumed from the start, δ_B is the correct phase shift. The Schrödinger equation (for s waves) for a linear dispersion reads

$$[-(i/\rho)(d/dx) + V(x)]\Psi(x) = (E - E_F)\Psi(x), \tag{37}$$

and the solution is given by

$$\Psi(x) = \exp\left[ikx - i\rho \int_0^x V(x) dx\right], \tag{38}$$

so that

$$\Psi(x) \sim e^{ikx + i\delta} \quad \text{as} \quad x \to \infty, \tag{39}$$

with

$$\delta = -\rho \int_0^\infty V(x) dx = -\pi\rho V \tag{40}$$

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[6] To evaluate an expression of the form $\langle e^A e^B \rangle$ [compare (32)] one makes use of the well-known relations $e^A e^B = e^{A+B+(1/2)[A,B]}$, which holds if $[A,B]$ is a c number, and $\langle e^{L(b^\dagger, b)} \rangle = \exp\frac{1}{2}\langle L^2(b^\dagger, b) \rangle$, where L is any linear combination of Bose operators.

[7] According to the Tomonaga model, $k_{\max} \approx \frac{1}{2} k_f$.

[8] J. M. Luttinger, J. Math. Phys. 4, 1154 (1963).

for $V(x)=2\pi V\delta(x)$, which has been used in the calculations above.

If one wants to stick to a more realistic model with a finite energy band, there are different ways to improve our result. The Luttinger model indicates that the incomplete result is in part due to the linear energy-momentum dispersion. A more realistic, namely, the quadratic dispersion leads to anharmonic terms, as can be seen from Schick's[9] paper. Another way would be to use the energy as variable instead of the momentum. But while this leaves the kinetic-energy term simple, the potential energy V would be very difficult to handle, again leading to anharmonic terms. An important point any way is to introduce a more realistic potential of arbitrary strength. But whatever one tries for larger V, one is soon struck with anharmonic effects. This should be sufficient to elucidate the situation. As there are other ways to calculate the exact transition rate,[1,10] we did not try to solve the anharmonic-oscillator problem.

[9] M. Schick, Phys. Rev. **166**, 404 (1968).

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Theory of Stimulated Raman Scattering*

CHEN-SHOW WANG†
Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92037
(Received 13 January 1969)

The semiclassical theory of spontaneous Raman scattering is reviewed. From the semiclassical theory we identify the parameters and dynamical variables involved in Raman scattering. A classical theory of stimulated Raman scattering is constructed. It is an extension of the theory of Shen and Bloembergen. We show that the system is a weakly coupled system. Because the driving term contains dynamical variables, the linewidth of the stimulated Stokes wave should be smaller than that of the spontaneous Stokes wave. Using Riemann's method, we obtain Kroll's solution rigorously, in a more general form. The steady-state limit is also derived rigorously. The conditions for the transient and steady-state gains are discussed. It is shown that for the transient case one may have an abrupt change of the Stokes gain versus incident laser power.

I. INTRODUCTION

WHEN a light beam passes through a medium, the most effective entities in scattering the light are electrons. The nuclear motion can modify the scattering of light by electrons. This leads to Raman[1] and Brillouin[2] scattering, with the scattered light shifted by the characteristic frequencies of the nuclear motion (optical and acoustical phonon frequencies for Raman and Brillouin scattering, respectively). If the intensity of the incident light beam is very high, the initially scattered waves can enhance further scattering of the incident wave, and lead to an exponential growth of the total scattered wave. This further scattering, enhanced by the initially scattered wave, is called stimulated scattering. In the past few years stimulated Raman scattering has been one of the most interesting topics in the field of nonlinear optics both experimentally and theoretically.[3] It is now clear that in order to have an appreciable stimulated Raman scattering, one requires the incident laser power to be at least several megawatts. For comparison with the experimental results, one usually assumes a steady-state spatial gain for the exponential growth, and good qualitative agreement is found.[3] There are several remarkable features in stimulated Raman scattering. The stimulated Stokes waves are emitted in the forward or backward directions with linewidths much smaller than the spontaneous linewidth. The phase-matching conditions require the stimulated anti-Stokes waves of different orders to be emitted in different coaxial cones.[4] Also, in many experiments, it is found that the Stokes gain of the strongest Raman line is anomalously high

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† Present address: Division of Engineering and Applied Physics, Harvard University, Cambridge, Mass.

[1] C. V. Raman, Indian J. Phys. **2**, 387 (1928). Raman scattering is an inelastic scattering of light, in which the scattered light is shifted by the frequency of a vibrational, rotational, or electronic excitation. In this paper we shall be concerned only with the vibrational excitation. The scattered light with frequencies shifted down are called the Stokes lines; those with frequency shifted up are called the anti-Stokes lines.

[2] L. Brillouin, Ann. Phys. (Paris) **17**, 88 (1922). Inelastic scattering of light, in which the frequency shift is the frequency of an acoustic phonon, is called Brillouin scattering.

[3] For a general review see N. Bloembergen, Am. J. Phys. **35**, 989 (1967).

[4] R. Y. Chiao and B. P. Stoicheff, Phys. Rev. Letters **12**, 290 (1964); E. Garmire, Phys. Letters **17**, 251 (1965).