# Electronic contribution to the thermal conductivity of noble metals in the low-temperature region ( $T \le 15$ )

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The electronic contribution to the low-temperature thermal conductivity of the noble metals copper and silver is calculated by a variational method for two different pseudopotentials. We describe the Fermi surface of the noble metals by the simple eight-cone model and take into account two OPW corrections which are very important at the lowest temperatures. The trial distribution function is expanded to the fifth power in the energy. Cubic terms in the energy are found to lower the first-order result by about 20% whereas the fifth-order corrections are negligible. The results of the calculation are discussed and compared with experimental data.

#### I. INTRODUCTION

Much work has been accomplished in recent years on the transport properties of simple metals, particularly potassium, in the low-temperature region  $(T \ll \Theta_p)$ . These works have shown the importance of umklapp processes, phonon drag, and the necessity of using a reliable trial function for a correct evaluation of the transport properties. On the other hand much less work has been devoted to the transport properties of noble metals for which additional complications arise due to the nonsphericity of the Fermi surface and the difficulty in obtaining a good pseudopotential for evaluating the electron-phonon matrix elements. A two-orthogonal-plane-wave (OPW) calculation of the electrical resistivity of noble metals has been carried out by Borchi et al.<sup>1</sup> using several pseudopotentials, a simple Debye phonon spectrum, and the eight-cone model to take into account the nonsphericity of the Fermi surface. Following similar lines Brett and Black<sup>2</sup> have evaluated the electrical and thermal resistivity of copper in the 1-20-K range. However, while taking into account the lattice dynamics by means of a Born-von Karman method, they perform the integration on the Fermi surface by evaluating the inverse relaxation time  $1/\tau_0$  at a particular azimuthal angle  $\theta$ and using this value for all  $\theta$  in the integration. This approximation is justified from the fact that  $1/\tau_0$  varies by no more than  $\pm 10\%$  as  $\theta$  is varied over the cone.<sup>2</sup>

In this paper we report the results of a two-OPW variational calculation on the electronic thermal conductivity of the noble metals Cu and Ag using the same model as in Ref. 1 both for the Fermi surface and the lattice dynamics. Since it is ex-

pected that phonon drag has a negligible influence on the thermal resistivity,<sup>3,4</sup> a comparison between theory and experiments is more significant for this transport property than in the electricalresistivity case, where a phonon-drag contribution may be of significant magnitude at low temperatures.

In the variational expression of the distribution function we consider only the energy dependence which has been shown to be of predominant importance with respect to the angular dependence in several previous calculations on the thermal conductivity of potassium.<sup>5,6</sup> It is plausible that this result may also apply to the noble metals. In Sec. II the relevant expressions for the calculation of the thermal conductivity are reported together with first-order variational results. Umklapp and normal contributions are separately presented in the 1–15-K temperature range. The calculations have been carried out using two different pseudopotentials; changes in the pseudopotential form factor have been seen to greatly influence the magnitude of the thermal resistivity. In Sec. III higher-order corrections to the variational calculation are evaluated by expanding the trial function up to the fifth order in the energy. Cubic terms are found to decrease the thermal resistivity by about 20% of the first-order variational result. On the other hand, fifth-order corrections are almost negligible. The results of the calculation are then discussed and compared with experimental data.

## **II. FIRST-ORDER VARIATIONAL CALCULATION**

If we take the phonon system to be in equilibrium,<sup>7</sup> the variational expression for the thermal resistivity W of a metal due to electron-phonon scattering is given by<sup>8</sup>

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$$W = \frac{1}{k_B} \int \int \int \left[ \Phi(\vec{k'}) - \Phi(\vec{k}) \right] P_{\vec{q}\vec{k}}^{\vec{k}'} d\vec{q} d\vec{k'} / \left| \int \vec{\nabla}_{\vec{k}} \epsilon_{\vec{k}} \Phi(\vec{k}) \frac{\partial f_{\vec{k}}}{\partial \epsilon_{\vec{k}}} d\vec{k} \right|^2,$$
(2.1)

where  $P_{\vec{q},\vec{k}}^{\vec{r}}$  is the probability for an electron of being scattered from  $\vec{k}$  to  $\vec{k'}$  by a phonon of wave vector  $\vec{q}$  ( $\vec{k} - \vec{k'} = \vec{q} + \vec{G}$ );  $\epsilon_i$  and  $\vec{v}_k$  are, respectively, the energy (measured relative to the local chemical potential) and the group velocity of an electron with wave vector  $\vec{k}$ . The trial function represents the deviation of the electron distribution function from the equilibrium distribution  $f_0$ .

As a first approximation, the trial function  $\Phi$  has

been chosen in the standard form

$$\Phi \propto \vec{v}_{t} \cdot \vec{u} \epsilon_{t}, \qquad (2.2)$$

where  $\vec{u}$  is a unit vector in the direction of the heat current. This differs from the form  $\Phi$  $\propto \vec{k} \cdot \vec{u} \in_{\vec{k}}$  used for the spherical Fermi-surface calculation. With this trial function and a local-pseudopotential approximation, the thermal electronic resistivity is

$$W = \left(\frac{1}{4\pi^{3}\hbar}\right)^{2} \frac{\pi m_{\text{opt}}^{2}}{6\hbar n^{2} e^{2} L_{0} T} \int \frac{dS_{\vec{k}}}{|\vec{\nabla}(\vec{k})|} \frac{dS_{\vec{k}'}}{|\vec{\nabla}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^{2} f_{s}(x) \\ \times \left[|\vec{\nabla}(\vec{k}) - \vec{\nabla}(\vec{k}')|^{2} - \frac{1}{2\pi^{2}} |\vec{\nabla}(\vec{k}) - \vec{\nabla}(\vec{k}')|^{2} x^{2} + \frac{3}{\pi^{2}} x^{2} |\vec{\nabla}(\vec{k})|^{2}\right],$$
(2.3)

where the double surface integrals are over the effective Fermi surface,  $g_{\lambda}(\vec{k},\vec{k'})$  is the electronic scattering matrix element, and  $f_s(x)$  is a statistical factor. Detailed expressions for  $g_{\lambda}$  and  $f_s(x)$  are reported in Ref. 1.  $L_0$  is the Lorenz number for free electrons.

As appears evident from Eq. (2.3), a detailed knowledge of the lattice dynamics and the Fermi surface is a prerequisite for a correct evaluation of the thermal conductivity. On the other hand, the use of a very accurate  ${\bf Fermi}\ {\bf surface}^9$  and a detailed lattice dynamics would considerably increase computation. In order to make the computational work tractable we will follow the same approach as in Ref. 1, i.e., we will consider the simple "eight-cone" model of the Fermi surface and a simplified description of the lattice dynamics by means of the Debye spectrum. It is likely that these approximate features of the calculation will in some way affect the results. On the other hand, in order to obtain more reliable results. an electronic-states expansion using more than two OPW's, especially at the top of the necks on the Fermi surface, should be necessary; this will require a great amount of computational work. In this paper we will consider in particular the relevance of changes in the pseudopotential form factor and only test the sensitivity of the thermalresistivity calculation with respect to small changes in the parameters of the eight-cone model and the Debye spectrum. In the eight-cone model the Fermi surface is made dependent on only the parameter r/p, i.e., the neck radius relative to the axis of the cone. In this calculation the same values as in Ref. 1 have been used for the longitudinal  $v_{L}$  and transverse  $v_{T}$  phonon velocities and the r/p quantities. More recent values,  $m_{opt}^{Cu} = 1.42 m_{free}$  (Ref. 10) and  $m_{opt}^{Ag} = 0.85 m_{free}^{,11}$ have been chosen for the optical-electron masses.

The integration in Eq. (2.3) was performed by means of the Gauss method with the help of a CDC 7600 computer. For each value of the temperature ture *T* the numerical evaluation of a three-dimensional integral in *q* space is required; this threedimensional integral is reduced to a series of onedimensional integrals by using the same procedure as in Ref. 1. Thirty Gauss points for each one-dimensional integral were used for T > 4 K, whereas forty points were necessary to obtain good convergence for  $T \le 4$  K.

The normal and umklapp processes have been evaluated separately in the calculation. In this respect we must mention that when a description of the electron states by multiple OPW's is adopted, this separation is not as trivial as it is for the one-OPW description. For our separation we used the Lawrence and Wilkins prescription.<sup>12</sup>

In Figs. 1(a) and 1(b) the first-order variational result for the thermal resistivity of Cu and Ag is reported. The resistivities are evaluated using two different pseudopotentials: a recent version of the Moriarty<sup>13</sup> pseudopotential and the Nand etal.<sup>14</sup> pseudopotential. From these figures it appears evident that the umklapp contribution exceeds the normal one by about one order of magnitude for the Moriarty pseudopotential and by a factor of 2 to 3 for the Nand *et al.* pseudopotential. On the other hand, the longitudinal and transverse umklapp components [not separately shown, in Figs. 1(a) and 1(b)] are of comparable magnitude. Moreover, contrary to what occurs in the case of potassium, the umklapp contribution does not freeze out at the lowest temperatures. These results are not surprising since the Fermi surface of the noble metals just touches the zone boundary at the (111) faces. We also see that the choice of the pseudopotential form factor significantly affects the magnitude and, to a lesser degree, the

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FIG. 1. (a) Temperature dependence of the thermal resistivity of copper calculated with the standard trial function given by Eq. (2.2) using the Moriarty pseudopotential (----) and the Nand *et al.* pseudopotential (----). In each case the total thermal resistivity has been explicitly separated into normal and umklapp components. The results obtained with the three different dielectric functions (see text) were so nearly identical that they could not be resolved on this scale. (b) Temperature dependence of the thermal resistivity of silver calculated with the standard trial function given by Eq. (2.2) using the Moriarty pseudopotential (-----) and the Nand et al. pseudopotential (----). In each case the total resistivity has been explicitly separated into normal and umklapp components. The results obtained with the three different dielectric functions (see text) were so nearly identical that they could not be resolved on this scale.

temperature dependence of the thermal resistivity, leading to changes of a factor of nearly 2. These calculations were repeated using several dielectric functions from Hartree,<sup>15</sup> Geldart and Vosko,<sup>16</sup> and Singwi *et al.*,<sup>17</sup> with no substantial variation in the thermal resistivities. Finally, in order to test the sensitivity of these results to the fine details of the Fermi surface, we repeated our calculations with new values of the r/p parameters differing from the previous ones by a small percentage (3%-6%). As in the electrical-resistivity case, no significative variation in the thermal resistivities of copper and silver was observed. Moreover, to test if the use of the simple Debye spectrum significantly affects the results, we subjected the  $v_L$  and  $v_T$  velocities to a  $\pm 10\%$ variation. The subsequent changes in the results should represent a useful estimate of the changes which could be produced by using a more realistic phonon spectrum in the calculation. The resulting changes lie in the range  $\mp 10\% - \mp 25\%$ . These are of the same magnitude as the changes observed by Leavens<sup>18</sup> in his calculation of the thermal resistivity of potassium, in which he used two different phonon spectra, one obtained from a fit to neutron data and the other from a first-principles calculation.

### III. HIGHER-ORDER VARIATIONAL CORRECTIONS TO THERMAL RESISTIVITY

Using a higher-order variational trial function in the energy, Sondheimer<sup>19</sup> first showed that corrections to the first-order variational result are important in the thermal-resistivity case. More recently Ekin<sup>20</sup> carried out a variational calculation for potassium, keeping terms up to cubic order and showed that the corrections are slightly less than 20% in the low-temperature regime where umklapp processes are frozen out. However, as umklapp processes become important. the corrections rise to slightly less than 30% near 5 K before diminishing at still higher temperatures. In other words higher-order corrections are significantly greater for the umklapp component than for the normal one. A similar variational calculation for potassium, expanding the trial function up to the seventh power in energy, has been carried out by Jumper and Lawrence.<sup>6</sup> In this work the seventh-order terms improve the results in such way that the exact (except for the negligible angular-dependent corrections) Leavens' clean  $limit^{18}$  is nearly attained. This work also attempts to evaluate the pure limit of the thermal resistivity of copper and silver by taking into account higher-order terms in the energy dependence of the trial function.

To estimate the magnitude of higher-order corrections, we expand the trial function up to the fifth power in the energy:

$$\Phi(\vec{k}) = \sum_{i} \alpha_{i} \varphi_{i}$$
$$\equiv \epsilon_{\vec{k}} \vec{v}_{\vec{k}}^{*} \vec{u} \left[ \alpha_{1}(T) + \frac{\alpha_{3}(T)}{(kT)^{2}} \epsilon_{\vec{k}}^{2} + \frac{\alpha_{5}(T)}{(kT)^{4}} \epsilon_{\vec{k}}^{4} \right].$$
(3.1)

The thermal conductivity is then obtained according to the usual variational procedure<sup>8</sup> by evaluating the expression

$$\kappa = \sum_{ij} X_i (P^{-1})_{ij} X_j.$$
 (3.2)

 $P_{ii}^{-1}$  denotes the *ij*th element of the 3 × 3 matrix  $P^{-1}$ . The matrix elements  $P_{ij} = \langle \varphi_i, P\varphi_j \rangle$  and  $X_i$  $=\langle \varphi_i, X \rangle$ , where the expressions for P and X are given in Ref. 8, are reported in the Appendix. In Tables I and II the results obtained for various orders of the trial-function expansion are presented for both the Moriarty and the Nand et al. pseudopotentials. Since for the noble metals the umklapp contribution is not frozen out at the lowest temperatures, it is not possible to see how much the higher-order corrections affect the umkalpp contribution with respect to the normal one. However, the third-order correction is seen to range from slightly more than 20% at 1.5 K to slightly less than 20% at 15 K (with a weak dependence on the particular pseudopotential and on the particular metal). The fifth-order correction is, on the other hand, practically negligible in

the whole range of temperatures, as it amounts at most to 0.3-0.4% of the total contribution. Also reported in Table I are the first-order variational results previously obtained for copper by Brett and Black,<sup>2</sup> who used the first version of the Moriarty pseudopotential<sup>21</sup> and the eight cone as a model of the Fermi surface. The differences with our first-order results are both in the absolute magnitude and in the temperature behavior of the thermal conductivity. In order to test if these differences can be due to differences in the peudopotentials, we have repeated our first-order calculation using the earlier version of the Moriarty pseudopotential<sup>21</sup>; however, no substantial variation is obtained with respect to our results reported in column III of Table I. From this test we can conclude that, whereas the differences in the absolute magnitude of the thermal conductivity can be plausibly ascribed either to the use of a different constant multiplicative factor in Eq. (2.3)or to a different extrapolation of the Moriarty pseudopotential<sup>22</sup> for  $q > 2k_F$ , the different temperature behavior must be ascribed to the different approach in treating the phonon spectrum and, perhaps, to the different integration procedure. In Figs. 2(a) and 2(b) the calculated total electronic contribution to the thermal resistivities is reported together with the recent experimental data for copper and silver by Rumbo.<sup>23</sup> The experimental data on a ultrapure sample by Ehrlich and Schriempf (ES)<sup>24</sup> and the data of Van Baarle et al.<sup>25</sup> are also reported for silver. Rumbo fitted his experimental data to a simple power law

TABLE I. Electronic components of the thermal resistivity of copper divided by the square of the temperature.  $W_1$  is the first-order,  $W_3$  the third-order, and  $W = W_1 + W_3 + W_5$  the total variational contribution, including the fifth-order correction. The results obtained with the Moriarty and the Nand *et al.* pseudopotentials are reported. Also reported are the first-order variational results by Brett and Black (Ref. 2). The units are  $10^{-6}$  cm/wattK.

T	Moriarty pseudopotential			Nand <i>et al.</i> pseudopotential			Brett and Black	
(K)	$W/T^2$	$W_1/T^2$	$-W_3/T^2$	$W/T^2$	$W_1/T^2$	$-W_{3}/T^{2}$	$W_1/T^2$	
1.5	78.50	98.00	19.50	35.24	44.19	8.95		
2.0	82.76	102.09	19.33	36.98	45.84	8.86		
2.5	86.62	105.87	19.25	38.31	47.07	8.76		
3.0	88.34	107.45	19.11	38.73	47.27	8.54	55.0	
4.0	89.68	108.50	18.79	38.03	46.45	8.42		
5.0	90.32	108.92	18.59	37.75	46.11	8.36	57.0	
6.0	90.77	109.02	18.22	37.32	45.64	8.32		
7.0	90.89	108.78	17.83	36.84	45.14	8.30	60.0 (7.5 K)	
8.0	90.81	108.46	17.56	36.39	44.68	8.29		
9.0	90.44	107.80	17.24	35.89	44.15	8.24		
10.0	89.91	107.09	17.01	35.40	43.64	8.21	65.0	
11.0	89.19	106.22	16.83	34.89	43.10	8.16		
12.0	88.33	105.30	16.72	34.39	42.56	8.10		
13.0	87.40	104.35	16.69	33.91	42.01	8.04		
14.0	86.38	103.35	16.68	33.40	41.44	7.96		
15.0	85.40	102.30	16.67	32.92	40.85	7.87		

Т	Mori	arty pseudop	otential	Nand <i>et al.</i> pseudopotential			
(K)	$W/T^2$	$W_{1}/T^{2}$	$-W_3/T^2$	$W/T^2$	$W_1/T^2$	$-W_{3}/T^{2}$	
1.5	77.86	95.01	17.15	25.08	30.79	5.71	
2.0	78.92	96.12	17.20	25.31	30.97	5.66	
2.5	79.83	97.10	17.27	25.49	31.10	5.60	
3.0	80.04	97.39	17.35	25.40	31.00	5.60	
4.0	80.03	97.57	17.55	25.16	30.76	5.59	
5.0	79.62	97.23	17.62	24.90	30.48	5.58	
6.0	79.08	96.85	17.80	24.62	30.21	5.57	
7.0	78.28	96.18	17.93	24.32	29.89	5.56	
8.0	77.20	95.35	18.04	24.00	29.54	5.52	
9.0	76.09	94.32	18.08	23.68	29.15	5.46	
10.0	74.90	93.10	18.02	23.32	28.71	5.36	
11.0	73.62	91.70	17.85	22.95	28.23	5.25	
12.0	72.23	89.97	17.52	22.58	27.70	5.10	
13.0	70.76	88.05	17.06	22.14	27.12	4.94	
14.0	69.25	86.01	16.52	21.75	26.53	4.76	
15.0	67.69	83.88	15.93	21.29	25.91	4.58	

TABLE II. Electronic components of the thermal resistivity of silver divided by the square of the temperature.  $W_1$  is the first-order,  $W_3$  the third-order, and  $W = W_1 + W_3 + W_5$  the total variational contribution, including the fifth-order correction. The results obtained with the Moriarty and the Nand *et al.* pseudopotentials are reported. The units are  $10^{-6}$  cm/wattK.



FIG. 2. (a) Temperature dependence of the thermal resistivity of copper evaluated taking into account fifth-order variational corrections. Solid lines are our calculated results: upper curve, the Moriarty pseudopotential; lower curve, the Nand *et al.* pseudopotential. The dashed line refers to the experimental behavior as obtained by Rumbo (Ref. 21) (for  $\rho_0 = 0.4 \times 10^{-9} \Omega$  cm). The results obtained with the three different dielectric functions were so nearly identical that they could not be resolved on this scale. (b) Temperature dependence of the thermal resistivity of silver evaluated taking into account fifth-order variational corrections. The solid lines are our calculated results: upper curve, the Moriarty pseudopotential; lower curve, the Nand *et al.* pseudopotential. The dashed line refers to the experimental behavior as obtained by Rumbo (Ref. 21) (for  $\rho_0 = 0.4 \times 10^{-9} \Omega$  cm). Other experimental data are from Refs. 22 (O) and 23 (D) (for  $\rho_0 = 0.88 \times 10^{-9} \Omega$  cm). The results obtained with the three different dielectric functions were so nearly identical that they could not be resolved on this scale.

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 $W(T) = BT^n$  with n = 1.6 (CU) and 1.7 (Ag) below 8.5 K. The measurements by ES on very pure silver show, on the other hand, a mean temperature dependence proportional to  $T^2$  over the range from about 4-12 K, in perfect agreement with the Bloch law. The data of Van Baarle *et al.* substantially agree with the Rumbo results. All these experimental data lie very close to the pure limit.<sup>23</sup>

Calculated thermal resistivities, on the other hand, do not follow a well-defined power law, but if one assumes the validity of a simple relation  $W \propto T^n$ , the exponent varies continuously, ranging from n = 2.3 (Cu) and n = 2.05 (Ag) at T = 2 K to n = 1.66 (Cu) and n = 1.67 (Ag) at T = 15 K, using the Moriarty pseudopotentials, and from n = 2.16 (Cu) and n = 2.03 (Ag) at T = 2 K to n = 1.79 (Cu) and n = 1.69 (Ag) at T = 15 K, using the Nand *et al.* pseudopotentials. Note also in Fig. 2(b) that the theoretical results, using the Nand pseudopotential, are lower than some experimental data. This contradicts the variational principle which should give values larger than the experimental ones. A simple explanation of this fact may be found in the inadequacy of the pseudopotential form factor. However, do not forget that the absolute magnitude of the thermal resistivity is also considerably sensitive to the multiplicative parameters, such as the optical mass, which appear in Eq. (2.3). From this comparison we can draw the conclusion that there are some discrepancies between the experiments and our calculations both in the absolute magnitude and the temperature dependence.

Some discrepancies can certainly be ascribed to the approximations in the shape of the Fermi surface and in the phonon spectrum, but the largest one arises from the uncertainties in the pseudopotential form factor which, of themselves, can explain the difference between the absolute magnitude of the experimental and theoretical results.

Finally, in this work higher-order variational corrections have been seen to contribute about 20% at low temperatures for the noble metals; more-over, umklapp scattering has been shown to give the dominant contribution to the thermal resistivity down to the lowest temperatures.

In conclusion, we briefly discuss a very recent calculation by Kus<sup>26</sup> on the electrical and thermal resistivities of copper and silver in the range 10-100 K. Kus, using the Leavens' approach,<sup>18</sup> numerically solves the Boltzmann transport equation for the energy dependence of the electron distribution function, assuming the lowest-order angular dependence. A comparison of our results with those obtained by Kus is, however, not possible, as they are for different temperature regions. However, since Kus uses the wrong Fermi surface (spherical) for the noble metals, his calculations will be poor at low temperatures.<sup>12</sup>

### APPENDIX

In this appendix we present, for the sake of completeness, the evaluated expressions of the matrix elements  $P_{ij} = \langle \varphi_i, P\varphi_j \rangle$  and  $X_i = \langle \varphi_i, X \rangle$ :

$$P_{11} = C(T)\pi^{2} \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k}')|} \frac{dS_{\vec{k}'}}{|\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^{2} f_{s}(x) \left(\frac{1}{3} |\vec{v}(\vec{k}') - \vec{v}(\vec{k}')|^{2} - \frac{1}{6\pi^{2}} |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^{2} x^{2} + \frac{1}{\pi^{2}} x^{2} |\vec{v}(\vec{k}')|^{2} \right),$$
(A1)  
$$P_{33} = C(T)\pi^{6} \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k}')|} \frac{dS_{\vec{k}'}}{|\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^{2} f_{s}(x) \left[\frac{31}{3} |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^{2} - |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^{2} \left(\frac{1}{140} \frac{x^{6}}{\pi^{6}} - \frac{7}{30} \frac{x^{2}}{\pi^{2}}\right) + |\vec{v}(\vec{k}')|^{2} \left(\frac{63}{10} \frac{x^{6}}{\pi^{6}} + \frac{21}{5} \frac{x^{2}}{\pi^{2}} + 2\frac{x^{4}}{\pi^{4}}\right)\right],$$
(A2)

$$P_{55} = C(T)\pi^{10} \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k}')|} \frac{dS_{\vec{k}'}}{|\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^2 f_{s}(x) \left[ \frac{2555}{33} |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^2 - |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^2 \right] \\ \times \left( \frac{1}{2772} \frac{x^{10}}{\pi^{10}} - \frac{31}{63} \frac{x^4}{\pi^4} - \frac{127}{6} \frac{x^2}{\pi^2} \right) \\ + |\vec{v}(\vec{k'})|^2 \left( \frac{23}{126} \frac{x^{10}}{\pi^{10}} + \frac{10}{3} \frac{x^8}{\pi^8} + 28 \frac{x^6}{\pi^6} + \frac{15500}{126} \frac{x^4}{\pi^4} + \frac{635}{3} \frac{x^2}{\pi^2} \right) \right],$$
(A3)

$$P_{31} = P_{13} = C(T)\pi^{4} \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k}')| |\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^{2} f_{s}(x) \left[ \frac{7}{15} |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^{2} - |\vec{v}(\vec{k}')|^{2} \left( \frac{1}{20} \frac{x^{4}}{\pi^{4}} - \frac{1}{6} \frac{x^{2}}{\pi^{2}} \right) + |\vec{v}(\vec{k}')|^{2} \left( \frac{x^{2}}{\pi^{2}} + \frac{1}{2} \frac{x^{4}}{\pi^{4}} \right) \right], \qquad (A4)$$

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where

and

 $X_1 = B(T) ,$ 

$$P_{51} = P_{15} = C(T)\pi^{6} \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k}')|} \frac{dS_{\vec{k}'}}{|\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k}')|^{2} f_{s}(x) \left[\frac{31}{21} |\vec{v}(\vec{k}) - \vec{v}(\vec{k}')|^{2} \left(\frac{1}{42} \frac{x^{6}}{\pi^{6}} - \frac{1}{6} \frac{x^{4}}{\pi^{4}} - \frac{7}{6} \frac{x^{2}}{\pi^{2}}\right) + |\vec{v}(\vec{k}')|^{2} \left(\frac{1}{3} \frac{x^{6}}{\pi^{6}} + \frac{5}{3} \frac{x^{4}}{\pi^{4}} + \frac{7}{3} \frac{x^{2}}{\pi^{2}}\right) \right],$$
(A5)

 $P_{53} = P_{35} = C(T)\pi^8 \int \frac{dS_{\vec{k}}}{|\vec{v}(\vec{k})|} \frac{dS_{\vec{k}'}}{|\vec{v}(\vec{k}')|} \sum_{\lambda} |g_{\lambda}(\vec{k},\vec{k'})|^2 f_s(x) \left[\frac{127}{5} |\vec{v}(\vec{k}) - \vec{v}(\vec{k'})|^2\right]$ 

$$- \left|\vec{\mathbf{v}}(\vec{\mathbf{k}}) - \vec{\mathbf{v}}(\vec{\mathbf{k}'})\right|^{2} \left(\frac{1}{504} \frac{x^{8}}{\pi^{8}} - \frac{7}{60} \frac{x^{4}}{\pi^{4}} - \frac{341}{126} \frac{x^{2}}{\pi^{2}}\right) + \left|\vec{\mathbf{v}}(\vec{\mathbf{k}'})\right|^{2} \left(\frac{19}{84} \frac{x^{8}}{\pi^{8}} + \frac{8}{3} \frac{x^{6}}{\pi^{6}} + \frac{77}{6} \frac{x^{4}}{\pi^{4}} + \frac{155}{7} \frac{x^{2}}{\pi^{2}}\right)\right],$$
(A6)

$$X_2 = B(T) \frac{\eta}{5} \pi^2$$
, (A9)

$$X_3 = B(T) \frac{31}{7} \pi^4 , \qquad (A10)$$

where

(A7)

(A8)

$$B(T) = -(k_B T)^2 n \pi^2 / 3m_{\text{out}} .$$
 (A11)

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 $C(T) = (1/4\pi^3\hbar)^2\pi k_B^2 T^3/6\hbar$ .

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