

DETERMINATION OF THE ELECTRON-PHONON COUPLING CONSTANT FROM OPTICAL MEASUREMENTS

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Optical measurements above the Debye temperature are used to determine the electron-phonon coupling constant  $\lambda$  which appears in the expression for the renormalized electron mass on the Fermi surface and in the equation for the critical superconducting transition temperature. The determination is based on a simple relation between  $\lambda$  and the electron relaxation time, due to electron-phonon interaction above the Debye temperature, when the entire phonon spectrum is excited. The values of  $\lambda$  obtained by optical measurements and by other methods are compared for Pb, Sn, In, Al, Zn, Nb, V, Nb<sub>3</sub>Sn, and V<sub>3</sub>Ga.

1. The electron-phonon coupling constant is an extremely important microscopic characteristic of metals which determines the renormalized electron mass on the Fermi surface, the electronic specific heat at low temperatures, and, perhaps most important, the critical superconducting transition temperature.

We know<sup>[1]</sup> that the superconducting transition temperature  $T_C$  can be expressed by

$$T_c = \frac{T_D}{1.45} \exp \left\{ - \frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right\}, \tag{1}$$

where  $T_D$  is the Debye temperature associated with the mean phonon frequency,  $\mu^*$  is the Coulomb pseudopotential, and  $\lambda$  is the electron-phonon coupling constant.

Equation (1) shows that  $\lambda$  is one of the basic parameters in the expression for  $T_C$ . Its exact determination is therefore very important for the theory of superconductivity. The electron-phonon coupling constant is determined from the expression<sup>[1]</sup>

$$\lambda = 2 \int_0^{\omega_0} \alpha^2(\omega) g(\omega) \frac{d\omega}{\omega}, \tag{2}$$

where the integration extends over the entire phonon spectrum,  $g(\omega)$  is the density of phonon states,  $\omega$  is the maximum phonon frequency, and

$$\alpha^2(\omega) g(\omega) = \frac{1}{(2\pi)^2} \left( \int \frac{dS_k}{v_k} \right)^{-1} \sum_{\sigma} \int \frac{dS_k}{v_k} \times \int \frac{dS_{k'}}{v_{k'}} |M(k, k')|^2 \delta(\omega - \omega_{k-k', \sigma}). \tag{3}$$

Here the integrals containing  $dS_k$  extend over the entire Fermi surface,  $v_k$  is the electron velocity on the Fermi surface, and  $\omega_{k-k', \sigma}$  is the phonon frequency.

2. Certain kinetic characteristics of metals which are associated with the electron-phonon interaction are also ultimately expressed using integrals of  $\alpha^2(\omega)g(\omega)$ . It is an important circumstance that if the entire phonon spectrum contributes to the electron-phonon coupling constant because of virtual transitions, then the kinetic characteristics associated with scattering by real phonons are determined by the portion of the spectrum that is excited at a given temperature.

We can therefore expect an essential correlation between the electron-phonon coupling constant and the kinetic properties of a metal that are measured above the Debye temperature, when the entire vibrational spectrum of the crystal is excited. Indeed, as was pointed out by one of the present authors in<sup>[2]</sup>, a direct relation exists between  $\lambda$  and the linearly temperature-dependent resistivity. It was shown, specifically, that the variation of  $\lambda$ , and therefore of  $T_C$ , due to any defects is proportional to the variation  $\Delta\rho$  of the linear resistivity coefficient that results from the same defects:

$$\frac{\Delta T_c}{T_c} \approx \frac{\Delta\rho}{\rho} \ln \frac{T_D}{T_c}.$$

The existence of this relationship has been confirmed experimentally in<sup>[3]</sup>. A similar concept of the correlation between high-temperature resistivity and the electron-phonon coupling constant was utilized in<sup>[4]</sup> to determine Fermi liquid parameters for alkali metals.

Hopfield<sup>[5]</sup> has recently proposed the use of infrared optical measurements to determine the electron-phonon coupling constant. His argument is based on the fact that the Drude-Zener equation is valid for transition metals and alloys at frequencies below those of interband transitions. This equation relates the complex dielectric constant  $\epsilon$  to the concentration  $N$  of conduction electrons and to the effective frequency  $\nu_{ep}$  of electron-phonon collisions:

$$\epsilon = A - \frac{4\pi e^2 N}{m\omega(\omega - i\nu_{ep})}. \tag{4}$$

Here  $e$  and  $m$  are the charge and mass of a free electron;  $\omega$  is the angular velocity of the light. In<sup>[5]</sup> the relation between  $\nu_{ep}$  and  $\lambda$  for  $T > T_D$  is given by

$$\lambda = \frac{\hbar}{2\pi} \frac{\nu_{ep}}{k_B T}. \tag{5}$$

Here  $k$  is Boltzmann's constant and  $T$  is the temperature. The values of  $\lambda$  calculated by Hopfield for the two metals Nb and Mo were in reasonable qualitative agreement with known values. Where electron scattering by impurities is significant Hopfield proposed the use of the derivative of the static resistivity with respect to temperature and the value of  $N/m$  obtained from infrared optical measurements.

3. It should be noted that the electron-phonon coupling constant  $\lambda$  is very directly related to the decay time  $\tau_{ep}$ , averaged over the Fermi surface, or a quasiparticle state with momentum  $k$ ; for  $T > T_D$  this is given by<sup>[6]</sup>

$$\frac{\hbar}{\tau_{ep}} = \frac{2k_B T}{(2\pi)^2} \left( \int \frac{dS_k}{v_k} \right)^{-1} \sum_{\sigma} \int \frac{dS_{k'}}{v_{k'}} \int \frac{dS_{k''}}{v_{k''}} |M(k, k')|^2. \quad (6)$$

In the relaxation time approximation the classical effective frequency of electron-phonon collisions which appears in the formula for the static electric conductivity at high temperatures is related to  $\tau_{ep}$  very simply:<sup>1)</sup>

$$\nu_{ep}^{el} = 1/\tau_{ep}. \quad (6')$$

The optical frequency  $\nu_{ep}$  in (4) is expressed by a more complicated equation than (6'). In optics  $\hbar\omega > k_B T$  even in the infrared region, and important effects are associated with the quantum character of the electron-photon interaction.<sup>[7,8]</sup> Although calculations performed for a spherical Fermi surface<sup>[7]</sup> indicate that there is little difference between  $\nu_{ep}$  and  $\nu_{ep}^{cl}$  when  $T > T_D$ , experiments have shown<sup>[9,10]</sup> that  $\nu_{ep}/\nu_{ep}^{cl} \approx 1.3$ . Therefore we consider that  $\lambda$  is determined more accurately by using  $\nu_{ep}^{cl}$ . Then, as seen from (2), (3), (6), and (6'), a valid expression for  $\lambda$  is (5) with  $\nu_{ep}$  replaced by  $\nu_{ep}^{cl}$ .

We can calculate  $\nu_{ep}^{cl}$  as follows. It is assumed in first approximation that the effective frequency of electron-phonon collisions is  $\nu_{ep}^{cl} + \nu_{ed}$ , where  $\nu_{ed}$  is the frequency of collisions with defects. Then

$$\nu_{ep}^{cl} + \nu_{ed} = \frac{e^2 N}{m \sigma_{st}}, \quad (7)$$

$$\frac{\nu_{ed}}{\nu_{ep}^{cl} + \nu_{ed}} = \frac{R_{res}}{R}. \quad (8)$$

Here  $\sigma_{st}$  is the static conductivity, and  $R_{res}/R$  is the ratio between the residual resistivity and the resistivity at the temperature  $T$ . For pure metals we have  $\nu_{ed} \ll \nu_{ep}^{cl}$ , but for alloys these quantities are alike in order of magnitude. For  $T > T_D$  we obtain  $\nu_{ep}^{cl}$  from (7) and (8) if  $N/m$  is known.

As shown in<sup>[9,11]</sup>, we obtain  $N/m$  from optical experiments in the long-wave region regardless of the character of the skin effect.<sup>2)</sup> It has been shown experimentally<sup>[9]</sup> that in the cases of polyvalent metals at high temperatures the corrections for an anomalous skin effect are small (comprising only a few percent).

For pure metals we can in most instances neglect

<sup>1)</sup>The use of  $\tau_{ep}$  to give the effective frequency of electron-phonon collisions, rather than the transport relaxation time, which differs from  $\tau_{ep}$  in that the integral includes the factor  $(1 - \cos \theta)$ , where  $\theta$  is the angle between the vectors  $k$  and  $k'$ , results from the fact that in the equation for the conductivity the average of a product of functions is replaced by the product of the averages and the factor  $N_{opt}/m$  appears. In the deduction of the Drude-Zener equation averages are separated similarly.

<sup>2)</sup>For cubic metals  $N/m = 2(2\pi\hbar)^{-3} \int v dS$ . For noncubic metals we have the tensor

$$\left( \frac{N}{m} \right)_i = \frac{2}{(2\pi\hbar)^3} \int \frac{v_i^2}{v} dS.$$

For polycrystals  $Sp(N/m)_i$  assumes the role of  $N/m$ .

### Experimental and theoretical values of $\lambda$

	$T_c, ^\circ K$	$\lambda_{exp}$		$\lambda_{calc}$	
		Optical method	Superconductivity method	[13]	[14]
Pb	7.2	1.03 <sup>[9,10]</sup>	1.12	1.32–1.34	1.55
Sn	3.7	0.68 <sup>[9,10,15]</sup>	0.60	0.78–0.99	0.92
In	3.4	0.66 <sup>[9,10,16]</sup>	0.69	0.84–1.16	0.74
Al	1.15	0.27 <sup>*</sup> [9,10]	0.38	0.52–0.53	0.41
Zn	0.85	0.23 <sup>**</sup> [9,17]	0.38	0.27–0.42	0.41
Nb ***	9.2	0.68 <sup>[19]</sup>	0.82	—	—
V	5.2	0.62 <sup>**</sup> [20]	0.60	—	—
Nb <sub>3</sub> Sn	18.2	0.89 <sup>[20]</sup>	1.12 <sup>****</sup>	—	—
V <sub>3</sub> Ga	14.2	0.91 <sup>[19]</sup>	0.98 <sup>****</sup>	—	—

\*Measurements for Al were performed at room temperature, which is somewhat lower than  $T_D$ .

\*\*Values of  $\sigma_{st}$  for Zn and V were obtained from handbooks.<sup>[18,25]</sup>

\*\*\*For Nb the values of  $\lambda$  obtained by Hopfield from<sup>[20,21]</sup> were 0.74 and 0.93, respectively. However, the optical measurements were here performed on mechanically polished samples, thus yielding results that apply to cold-hardened metal.

\*\*\*\*These values of  $\lambda$  were obtained in<sup>[24]</sup> by a procedure similar to that used [1] with  $\mu^* = 0.13$ .

the constant  $A$  in (4) and use both  $Re \epsilon$  and  $Im \epsilon$  to determine  $N/m$ ; the corresponding equations are given in<sup>[9]</sup>. For alloys the contribution of virtual interband transitions in  $Re \epsilon$  is comparable with the contribution of conduction electrons; this means that  $A$  should not be neglected. Therefore only  $Im \epsilon$  should be used. A very convenient procedure for extracting  $N/m$  from the optical data for alloys has been described in<sup>[12]</sup>.

After  $\nu_{ep}^{cl}$  is obtained,  $\lambda$  is calculated from (5); the results are shown in the table. For all the metals and alloys except Zn and V the optical constants and static conductivity were measured for the same samples. The errors were 3–7% for pure metals and 20–25% for alloys.

Our table also contains values of  $\lambda_{exp}$  obtained by McMillan<sup>[1]</sup> from superconductivity data, and values of  $\lambda_{calc}$  calculated in<sup>[2,3,14]</sup> in the pseudopotential approximation. In<sup>[13]</sup> several values of  $\lambda$  corresponding to different pseudopotentials were obtained; the table gives the extreme values. We observe that for the non-transition metals the values of  $\lambda$  derived from optical measurements are in good agreement with those obtained by McMillan and with other values calculated by several authors. The values of  $\lambda$  are correlated with values of  $T_c$ .

The situation is not so good for the transition metals, where the values of  $\lambda$  exhibit very little correlation with each other or with  $T_c$ . This result appears to indicate that the transition-metal values are significantly influenced by band structure or anisotropy, which are not taken into account in (5).

For the alloys Nb<sub>3</sub>Sn and V<sub>3</sub>Ga we can use (1) to obtain  $\mu^*$ . Using  $T_D = 304^\circ K$  for Nb<sub>3</sub>Sn and  $310^\circ K$  for V<sub>3</sub>Ga, we obtain 0.06 and 0.11, respectively, as the values of  $\mu^*$ . The small values of  $\mu^*$  may indicate a phonon mechanism of superconductivity in these alloys.

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