# Resistivities and mean free paths in individual layers of a metallic multilayered structure

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There exists in the literature a number of transport measurements performed on metallic superlattices. In this paper a method is proposed which, given such measurements performed at two different temperatures, allows determination of resistivities and mean free paths (MFP's) of carriers in individual layers comprising a superlattice. The method is illustrated for Nb/Al (author's data) and Nb/Cu (data of Werner, Banerjee, Yang, Falco, and Schuller). The behavior of the MFP in individual layers as a function of layer thickness turns out to be both unexpected and instructive. In particular, we find that in all cases except for Cu in Nb/Cu, the MFP is significantly (factor of 2-3) smaller than the layer thickness. Previously, for the lack of such information, it was uniformly assumed that the MFP is limited by the layer thickness,  $L \sim d$ . Knowledge of the MFP is important for interpretation of various experiments on metallic superlattices, in particular for better understanding of superconducting proximity systems and tunneling measurements.

#### I. INTRODUCTION

Resistive measurements on metallic multilayered structures were reported in a number of papers.<sup>1-3</sup> One of the most intriguing aspects of these studies is that thin continuous metal layers can be grown on top of other metals with relative ease, while the equally thin single films grown on insulating substrates tend to form islands. In the case of multilayers, an obvious consequence, however, is the difficulty of knowing resistivities and mean free paths (MFP's) in individual layers from the measured resistivity of the whole stack. A typical resistivity  $\rho$  vs superlattice period  $\lambda = d_1 + d_2$  curve (obtained on Nb/Al multilayers in the author's laboratory) is shown in Fig. 1. In the limit of long periods resistivity falls between that of individual metals, and in the short-period limit resistivity approaches a typical metallic saturation value of  $\sim 200$  $\mu\Omega$  cm. The curve shown in Fig. 1, while bordering on triviality as far as physics is concerned, provides a useful check for the overall soundness of the data. For example, if individual metals were dirty, or if they were mixing together during the deposition, resistivity in the limit of long periods would have been higher than either one of the bulk resistivities of the constituents. More can be learned by plotting  $\rho$  as a function of  $\lambda^{-1}$ , as is often done in the studies of thin films, and as was done for multilayers in the work of Zheng et  $al.^1$  and Werner et  $al.^2$ In Refs. 1 and 2 it was found that there exists some range of thicknesses in which  $\rho \propto \lambda^{-1}$ . The implication is that in this range the MFP's  $L_1, L_2$  in individual layers are limited by the layer thicknesses  $d_1, d_2$ . Outside of this range MFP's are saturated on the small- $\lambda$  end, and limited by impurities and/or phonons inside the layers on the long- $\lambda$  end. Strictly speaking, one has no reason to claim that  $\rho \propto \lambda^{-1}$  implies  $L_1 \cong d_2$  and  $L_2 \cong d_2$ ; all one can say is that  $L_1 = \alpha_1 d_1$  and  $L_2 = \alpha_2 d_2$  with  $\alpha_1, \alpha_2 \le 1$ . We shall see that indeed in some cases  $\alpha$ 's can be considerably smaller than unity.

It appears that much more could be learned if we would try and find resistivities and MFP's in individual layers. By comparing the MFP with layer thicknesses one could then hope to learn more about the quality of the layers; the knowledge of the MFP would also be useful in interpreting experiments on superconducting layered structures, such as the one reported in Ref. 4. In addition, some of the more promising applications of layered structures have been found in tunneling spectroscopy<sup>5,6</sup> and in refractory Josephson junctions based on Nb/Al structures.<sup>7</sup> In these systems an electrode of a tunnel junction consists of a bilayer of a superconductor and a normal (or low  $T_c$ ) metal. The physical models which describe such systems require the knowledge of the MFP of electrons in



FIG. 1. Room-temperature resistivity of Nb/Al multilayers as a function of superlattice period  $\lambda$ .

(4)

normal layers. The latter is presently being treated as an adjustable parameter.<sup>5</sup> The analysis presented in this paper in principle allows one to perform independent resistivity measurements and deduce the MFP in a bilayered structure identical to the one used in tunneling.

In Secs. II and III we present certain assumptions which greatly simplify the problem of finding the MFP in individual layers, write down the basic equations, and give their solution in terms of measurable quantities. In Sec. IV we describe preparation of multilayered Nb/Al samples and reference films of Nb and Al; Sec. V is devoted to a question of finding the MFP from known resistivity; in Sec. VI we apply equations developed in Secs. II and III to the analysis of our data on Nb/Al, and the data of Werner *et al.*<sup>2</sup> on Nb/Cu. Section VII contains a discussion of the obtained relationship between layer thickness and the MFP in multilayers.

#### **II. THE CASE OF LOW RESISTIVITIES**

We will make the following assumptions.

(1) If the bulk MFP of electrons is longer than the layer thickness d, the MFP L in the layer is limited by the interfaces between the layers:  $L \leq d$ . Unfortunate as it may be for the observation of possible superlattice effects, this assumption is probably true for most metallic multilayers made today, with the notable exception of Nb/Ta superlattices<sup>8,9</sup> (see Ref. 10, however). This is the major simplification which allows one to treat individual layers as independent resistors connected in parallel. The limits of validity of this assumption constitute an interesting problem. One can argue that if scattering at the interfaces is entirely diffuse, then the fact that electrons are crossing from one layer into the other does not invalidate the assumption of parallel connections. Indeed, due to the condition of charge neutrality, the number of electrons entering into a given layer will have to be equal to the number of electrons leaving it. Each electron experiences diffuse scattering at the interface and loses any memory of the drift velocity which it acquired in the field. Hence it does not matter whether the layers are physically separated or not.

In the presence of some specular scattering events at the interfaces justification of the first assumption appears difficult; a more-detailed analysis would be required to see whether in this case it remains a reasonable approximation.

(2) The second assumption concerns the size of the temperature-dependent (phonon) part of resistivity in a layer, h(T). If resistivity is not too high (i.e., if  $L >> a_n$ , where  $a_n$  is the nearest-neighbor distance in the lattice), we can assume the validity of the Mattheissen's rule:

$$\rho(T) = \rho(0) + h^{(b)}(T) , \qquad (1)$$

where the residual resistivity  $\rho(0)$  includes all temperature-independent contributions, including interface scattering, and  $h^{(b)}(T)$  is the bulk temperature-dependent resistivity characteristic of a given material. In a more resistive (thinner) layer this assumption is no longer true. This case will be considered in Sec. III.

We have two metals of which the bi- or multilayered sample is composed with individual layer thicknesses  $d_1$ and  $d_2$ . Measurements are performed at room temperature (T=r) and at  $T \cong 0$   $(T \gtrsim T_c$  in superconductors). We know the following:

(1) The ratio of the layer thicknesses  $t = d_1/d_2$ .

(2) Resistivities of the multilayered sample  $\rho_m(r)$  and  $\rho_m(0)$ .

(3) Bulk thermal resistivities of both metals:  $h_1^{(b)} = \rho_1^{(b)}(r) - \rho_1^{(b)}(0)$  and  $h_2^{(b)} = \rho_2^{(b)}(r) - \rho_2^{(b)}(0)$ .

We want to find resistivities in individual layers:  $\rho_1(0)$ ,  $\rho_1(r)$ ,  $\rho_2(0)$ ,  $\rho_2(r)$ .

Consider a sample of unit length and width which is made of *n* double layers  $d_1+d_2$ , so that the total thickness is  $n(d_1+d_2)$ . The resistance  $R_m$  of such a sample is

$$R_m = \frac{\rho_m}{n(d_1+d_2)} \; .$$

The resistance of *n* layers of the type 1 is  $R_1 = \rho_1/nd_1$ and of the type 2 is  $R_2 = \rho_2/nd_2$ . Using our first assumption, we write  $R_m^{-1} = R_1^{-1} + R_2^{-1}$  and arrive at the equation relating resistivities

$$\frac{1+t}{\rho_m(T)} = \frac{t}{\rho_1(T)} + \frac{1}{\rho_2(T)} .$$
(2)

Note that in a homogeneous sample  $\rho_1(T) = \rho_2(T)$ , and Eq. (2) correctly gives  $\rho_m(T) = \rho_1(T) = \rho_2(T)$ . Substituting T = 0 and T = r into Eq. (2), we obtain two equations. Two more are obtained by writing Eq. (1) for each material at T = r. So we have four equations for four unknowns:

$$\rho_{1}(r) = \rho_{1}(0) + h_{1}^{(b)}(r) ,$$

$$\rho_{2}(r) = \rho_{2}(0) + h_{2}^{(b)}(r) ,$$

$$\frac{1+t}{\rho_{m}(0)} = \frac{t}{\rho_{1}(0)} + \frac{1}{\rho_{2}(0)} ,$$

$$\frac{1+t}{\rho_{m}(r)} = \frac{t}{\rho_{1}(r)} + \frac{1}{\rho_{2}(r)} .$$
(3)

This leads to a quadratic equation for any of the variables; solving first for  $\rho_1(0)$ , we obtain

$$\rho_1(0) = \frac{-b + (b^2 - 4ac)^{1/2}}{2a} ,$$
  
$$\rho_2(0) = \frac{\rho_1(0)\rho_m(0)}{(1+t)\rho_1(0) - t\rho_m(0)} ,$$

where

$$\begin{aligned} a &= (1+t) [\rho_m(r) - \rho_m(0) - (1+t)h_2^{(b)}], \\ b &= (1+t) \{ t [\rho_m(0) + \rho_m(r)] h_2^{(b)} \\ &+ [\rho_m(r) - \rho_m(0)] h_1^{(b)} - (1+t)h_1^{(b)} h_2^{(b)} \}, \\ c &= t \rho_m(0) [(1+t)h_1^{(b)} h_2^{(b)} - \rho_m(r)(th_2^{(b)} + h_1^{(b)})]. \end{aligned}$$

Knowing  $\rho_1(0)$  and  $\rho_2(0)$ , we obtain  $\rho_1(r)$  and  $\rho_2(r)$  from the first two equations of (3). Note that substituting  $h_1^{(b)} = h_2^{(b)}$  into (4) we correctly obtain  $\rho_1(0) = \rho_2(0)$  $= \rho_m(0)$ .

or

(5)

## III. THE GENERAL CASE: TAKING ACCOUNT OF RESISTIVITY SATURATION

As mentioned in the preceding section, the Mattheissen's rule (1) will fail to be a good approximation once L starts approaching  $a_n$ . As the MFP becomes shorter with the decreasing layer thickness, h(T) will become smaller than  $h^{(b)}(T)$  and, eventually, in the regime  $L \sim a_n$ , it will approach zero and even become negative,<sup>2</sup> the same way as it happens in alloys<sup>11</sup> and, generally, in all disordered metals. This fact is usually described as the relation between the temperature coefficient of resistivity and resistivity (the Mooij correlation<sup>11</sup>). The physics behind the Mooij correlation<sup>12</sup> attracted much attention in recent years; we do not intend to review this subject here. Let us just note that the appearance of h(T) < 0 in very thin layers is no more surprising than the Mooij correlation itself; we see no need for explanations designed specifically for layered structures, as was done in Ref. 3, where the interface properties were considered to be temperature dependent. For one thing, a sample made up of very thin (say, one monolayer thick:  $d_1 = d_2 \simeq 4$  Å) layers is indistinguishable from the mixture of the two components. Such mixture still has the negative h(T) (see Table I), even in the absence of interfaces.

In the regime where h(T) > 0, the decrease of h(T) for shorter MFP's (higher resistivity) is known as resistivity saturation; it can be described with the help of the "parallel-resistors" formula<sup>13</sup> (not to be confused with our assumption of layers as resistors in parallel):

$$\rho^{-1} = \rho_{id}^{-1} + \rho_s^{-1}$$

$$[\rho(0)+h(T)]^{-1}=[\rho_{id}(0)+h_{id}^{(b)}(T)]^{-1}+\rho_s^{-1},$$

where  $\rho$  is the total measured resistivity consisting of the residual and thermal parts  $\rho(0)$  and h(T),  $\rho_{id}(0)$  and  $h_{id}^{(b)}(T)$  are the "ideal" or Boltzmann residual and thermal resistivities, i.e., resistivities which would be observed if L could take on arbitrarily small values, and  $\rho_s$  is a constant. Several theoretical justifications of (5) now exist; Gurvitch<sup>14</sup> derived (5) in a particularly simple way by considering the statistical distribution of the free paths (times) between collisions coupled with the Mott-Ioffe-Regel<sup>15</sup> criterion which can be formulated as a statement that in a metal  $L \ge a_n$ . In this case  $\rho_s$  is simply a resistivity at  $L = a_n$ . In the free-electron approximation it can be estimated from the formula<sup>14,16</sup>

$$\rho_s = (3\pi^2)^{1/3} \hbar e^{-2} n^{-2/3} a_n^{-1}$$

or, substituting *n* in terms of the atomic weight *A*, density  $\gamma$ , and valence *z*:

$$\rho_s = \frac{178.1}{a_n} \left[ \frac{A}{\gamma z} \right]^{2/3} . \tag{6}$$

Here  $[\rho_s] = \mu \Omega \text{ cm}$ ,  $[a_n] = \dot{A}$ , [A] = atomic units, and $[\gamma] = g/\text{cm}^3$ . Using the valencies of 5, 3, and 1 for Nb, Al, and Cu we obtain from (6):  $\rho_s^{\text{Nb}} = 104 \ \mu \Omega \text{ cm}$ ,  $\rho_s^{\text{Al}} = 139 \ \mu \Omega \text{ cm}$ , and  $\rho_s^{\text{Cu}} = 257 \ \mu \Omega \text{ cm}$ .

System	λ (Å)	<i>d</i> <sub>1</sub> (Å)	$d_2$ (Å)	$ \rho_m(r) $ ( $\mu \Omega  \mathrm{cm}$ )	$ ho_m(0)$ ( $\mu\Omega \mathrm{cm}$ )	$ ho_1(r)$ ( $\mu\Omega$ cm)	$ ho_1(0)$ ( $\mu \Omega  \mathrm{cm}$ )	$ ho_2(r)$ ( $\mu \Omega \mathrm{cm}$ )	$ ho_2(0)$ ( $\mu\Omega \mathrm{cm}$ )	$C'_1$ ( $\mu\Omega \operatorname{cm} \operatorname{\AA}$ )	$C'_2$ ( $\mu\Omega\mathrm{cm}\mathrm{\AA}$ )
Nb film		4632				20.4	4.64	****		342	
Al film			4292					3.80	0.88		575
Nb(1)-Al(2)	375	187	188	11.7	5.25	20.77	5.14	8.16	5.37	342	575
	116.5	57.3	59.2	19.81	12.38	25.4	11.2	16.3	13.8	355	575
	73	37	36	41.1	30.90	35.15	22.9	49.7	48.0	430	575
	47	27	20	93.1	83.9	85.85	80.66	105.1	104.4	510	575
	27.1	13.4	13.7	197.9	207.7						
	8	4	4	205.8							
Nb(1)-Cu(2) <sup>a</sup>	401	200.5	200.5	10.28	5.57	19.96	5.9	6.92	5.27	345	851
	343	171.5	171.5	12.43	7.23	21.0	7.26	8.83	7.20	345	851
	189	94.5	94.5	16.46	11.27	24.6	11.81	12.37	10.78	355	851
	146	73.0	73.0	20.36	15.0	27.5	15.41	16.17	14.61	365	851
	74.3	37.15	37.15	35.64	30.02	39.62	29.10	32.39	30.98	476	851
	58.3	29.15	29.15	39.27	34.29	44.82	35.05	34.94	33.57	490	851
	48.8	24.4	24.4	49.0	43.57	49.65	40.53	48.36	47.1	500	851
	43.2	21.6	21.6	50.42	45.72	53.91	45.36	47.35	45.36	500	851
	36.7	18.35	18.35	56.91	52.89	61.98	54.48	52.61	51.39	505	851
	35.5	17.75	17.75	55.32	50.81	58.07	50.04	52.82	51.60	510	851
	31.4	15.7	15.7	58.56	54.55	63.0	55.64	54.7	53.51	510	851
	28.8	14.4	14.4	81.14	76.80	72.44	66.26	92.22	91.32	510	851
	22.7	11.35	11.35	90.3	88.46	102.34	99.21	80.79	79.81	510	851
	19.0	9.5	9.5	126.5	128.81						

TABLE I. Measured and calculated parameters of Nb/Al and Nb/Cu multilayers; saturation taken into account.

<sup>a</sup>Nb-Cu data of T. R. Werner, I. Banerjee, Q. S. Yang, C. M. Falco, and I. K. Schuller presented in a reduced form in Ref. 2. Absolute values of resistivity sent to me by I. K. Schuller.

(9)

Perhaps a more reliable way of estimating  $\rho_s$  is to use the results of band-structure calculations, as was done in Ref. 14 (also see below). Finally in some cases  $\rho_s$  can be obtained from the high-temperature fits of the resistivity to the formula (5).<sup>13</sup> Mattheissen's rule (1) follows from (5) in the limit  $\rho_s \rightarrow \infty$ , which is equivalent to  $a_n \rightarrow 0$ .

We can further write (5) for a bulk metal:

$$[\rho^{(b)}(0) + h^{(b)}(T)]^{-1} = [\rho^{(b)}_{id}(0) + h^{(b)}_{id}(T)]^{-1} + \rho^{-1}_{s}$$
(7)

in which case we know  $\rho^{(b)}(0)$  and  $h^{(b)}(T)$ . In many metals with a high resistance ratio the bulk residual resistivity  $\rho^{(b)}(0)$  can be neglected. In certain cases, however,  $\rho^{(b)}(0)$ may be quite large: for example, in the high- $T_c$  A-15 compounds. We can write (5) and (7) for each metal at T=0 and at T=r. At T=0 all thermal parts will drop out and we will obtain relations between the various residual resistivities. As previously mentioned, two equations are obtained at T=0 and T=r from (2), making a total of ten equations:

$$\frac{1+t}{\rho_m(0)} = \frac{t}{\rho_1(0)} + \frac{1}{\rho_2(0)} ,$$

$$\frac{1+t}{\rho_m(r)} = \frac{t}{\rho_1(r)} + \frac{1}{\rho_2(r)} ,$$

$$\frac{1}{\rho_j(0)} = \frac{1}{\rho_{id_j}(0)} + \frac{1}{\rho_{s_j}} ,$$

$$\frac{1}{\rho_j(r)} = \frac{1}{\rho_{id_j}(0) + h_{id_j}^{(b)}(r)} + \frac{1}{\rho_{s_j}} ,$$

$$\frac{1}{\rho_j^{(b)}(0)} = \frac{1}{\rho_{id_j}^{(b)}(0)} + \frac{1}{\rho_{s_j}} ,$$

$$\frac{1}{\rho_j^{(b)}(r)} = \frac{1}{\rho_{id_j}^{(b)}(0) + h_{id_j}^{(b)}(r)} + \frac{1}{\rho_{s_j}} ,$$
(8)

$$i = 1.2$$
.

The known (input) parameters are t,  $\rho_m(0)$ ,  $\rho_m(r)$ ,  $\rho_1^{(b)}(0)$ ,  $h_1^{(b)}(r)$ ,  $\rho_2^{(b)}(0)$ ,  $h_2^{(b)}(r)$ ,  $\rho_{s_1}$ , and  $\rho_{s_2}$ . The ten unknowns are  $\rho_1(0)$ ,  $\rho_1(r)$ ,  $\rho_2(0)$ ,  $\rho_2(r)$ ,  $\rho_{id_1}(0)$ ,  $\rho_{id_2}^{(b)}(0)$ ,  $\rho_{id_2}^{(b)}(0)$ ,  $\rho_{id_2}^{(b)}(0)$ ,  $h_{id_1}^{(b)}$ , and  $h_{id_2}^{(b)}$ .

Despite the lengthy appearance, these equations conceptually are as simple as the previous ones, differing only in the corrections to the Mattheissen's rule brought about by the short MFP and expressed through the "parallelresistor" formula. As previously, the solution reduces to a quadratic equation. The results are the following:

$$\begin{split} \rho_1(0) &= \frac{-B + (B^2 - 4AC)^{1/2}}{2A} ,\\ \rho_2(0) &= \frac{\rho(0)\rho_1(0)}{(1+t)\rho_1(0) - t\rho(0)} ,\\ \rho_j(r) &= \frac{\rho_{s_j}[\rho_{s_j} - \rho_j(0)(1-\rho_{s_j}h_j)]}{\rho_{s_j}[\rho_j(0)\rho_{s_j} + 1] - \rho_j(0)(1-\rho_{s_j}h_j)} ,\end{split}$$

where

$$h_j = \frac{\rho_j^{(b)}(r)}{1 - \rho_j^{(b)}(r) / \rho_{s_j}} - \frac{\rho_j^{(b)}(0)}{1 - \rho_j^{(b)}(0) / \rho_{s_j}}, \quad j = 1, 2$$

and

$$\begin{split} A = F[\alpha(1+t) + \rho_m(r)(t-q\alpha)] - \rho_m(r)\alpha E , \\ B = (1+t)(\rho_{s_1}h_1F - \alpha\rho_{s_2}h_2t\rho_m(0)) \\ -\rho_m(r)[\rho_{s_1}F(t+qh_1) - \alpha\rho_{s_2}t\rho_m(0)(1+qh_2) \\ + Eh_1\rho_{s_1} + t^2\rho_{s_2}h_2\rho_m(0)] , \\ C = \rho_{s_1}\rho_{s_2}t\rho_m(0)[-(1+t)h_1h_2 \\ +\rho_m(r)(th_2+h_1+qh_1h_2)] , \\ \alpha = \rho_{s_1} - h_1, \ \beta = \rho_{s_2} - h_2, \ q = \frac{t}{\rho_{s_1}} + \frac{1}{\rho_{s_2}} , \\ E = (1+t)\rho_{s_2} - \rho_m(0) , \\ F = \beta\rho_m(0) + \rho_{s_2}h_2(1+t) . \end{split}$$

It is easy to show that in the limit  $\rho_{s_1}, \rho_{s_2} \rightarrow \infty$ , these equations reduce to (4).

### **IV. EXPERIMENTAL**

Nb/Al multilayered structures with periods  $\lambda = d_1 + d_2$ from 8 to ~400 Å were deposited in a magnetron sputtering system as described in Refs. 17 and 18. The important difference between these and our previous<sup>17</sup> samples is in the use of a partition between the Nb and Al sputter guns. We found that without a partition resistivities of samples even with large layer thicknesses did not decrease below ~60  $\mu\Omega$  cm. This was found to be due to unforeseen mixing occurring during the deposition of the two metals; we estimate about 3–4 at.% of Al in Nb in the old samples. Preliminary x-ray examination of the new samples indicates better layering.<sup>19</sup>

Samples were deposited on 90° sapphire substrates. The four-probe geometry was defined with shadow masks. The rates were calibrated in separate runs and were correlated with the amount of weak (groove depth) on the sputtering target.<sup>20</sup> The calculated total thicknesses (usually ~3000 Å) of the multilayered samples obtained from the rate calibration were within  $\pm 6\%$  of the measured thicknesses. We estimate the error in absolute resistivities to be  $\pm (6-8)\%$ .

The substrates were not heated intentionally; however, sputtering produced a certain amount of heating. If a substrate was simply laid down on a substrate table, the temperature raised to about 200 °C and saturated after 5 min of steady Nb deposition at the power of 400 W. Of course, in multilayers the substrate table rotates, and a sample spends only a fraction of the total time under the gun, thus reducing the heating. Thus, samples were prepared without special provisions for cooling. Films of pure Nb and Al ~4000 Å thick were also deposited. The thermal parts of resistivities of Nb and Al were found to be 15.4 and 2.93  $\mu\Omega$  cm, respectively (see Table I). These values are within the claimed error from the pub-

lished bulk values of 14.5 and 2.74  $\mu\Omega$  cm.<sup>21</sup>

The data for Nb/Al and Nb/Cu multilayers is collected in Table I. The room-temperature resistivity  $\rho_m(r)$  of Nb/Al is plotted as a function of  $\lambda = d_1 + d_2$  in Fig. 1, the resistivities of bulk Nb and Al are indicated by arrows.

### **V. MFP ESTIMATES**

Before we go ahead with our analysis, let us discuss the ways of estimating the MFP from the known resistivity. Indeed, the MFP often is a more instructive quantity than resistivity, especially in our case, when we can compare the MFP with the layer thickness. From the Drude formula one finds a familiar expression

$$\rho L = \frac{mv_F}{e^2 n} = C' \; .$$

One way to find the free-electron value of the constant C' is to evaluate it in the saturation limit  $L = a_n$ ; then, from (6) we obtain

$$C' = \rho_s a_n = 178.1 \left[ \frac{A}{\gamma z} \right]^{2/3}.$$
 (10)

Using valencies of 1, 3, and 5 for Cu, Al, and Nb, we find free-electron values  $C'_{Cu} = 659 \ \mu\Omega \text{ cm Å}$ ,  $C'_{Al} = 397 \ \mu\Omega \text{ cm Å}$ , and  $C'_{Nb} = 298 \ \mu\Omega \text{ cm Å}$ . Of course we have no reason to hope that this simple estimate will indeed be accurate. Yet we shall see that it is not far off.

A better estimate can be obtained using the results of band-structure calculations of the Fermi-surface-averaged Drude plasma frequency  $\langle \Omega_p^2 \rangle^{1/2}$  and Fermi velocity  $\langle v_F \rangle$  through the relation<sup>22</sup>  $\rho = 4\pi / \langle \Omega_p^2 \rangle \tau$ . Writing  $\tau = L / \langle v_F \rangle$ , we find

$$\rho L = C' = \frac{4\pi \langle v_F \rangle}{\langle \Omega_{\rho}^2 \rangle} . \tag{11}$$

We note that, strictly speaking, C' is not a constant for a given material, despite the assertion to the contrary commonly found in the literature; at least it is not a constant in transition metals and compounds where the bandstructure parameters  $\Omega_p$  and  $v_F$  may be sensitive to the lifetime-broadening effects.<sup>23</sup> We plot  $C'[\rho(0)]$  for Nb in Fig. 2, using the results of Ref. 23 for  $\rho$ -dependent  $\langle v_F \rangle$  and  $\langle \Omega_{\rho}^2 \rangle^{1/2}$ . Note that if C' is  $\rho$  dependent, so is  $\rho_s$ . In clean Nb, we find  $C' = 350 \ \mu\Omega \ \text{cm A}$ . The obtained value of C' for bulk Nb is in good agreement with the value of  $\rho L = 375 \ \mu\Omega \,\mathrm{cm}\,\mathrm{\AA}$  deduced<sup>24</sup> in superconductive and magnetic studies of Nb. Our  $C'_{0}$  is also in reasonable agreement with  $C = 460 \ \mu\Omega \text{ cm} \text{ \AA}$  offered as a bandstructure value in Ref. 25. All these values, however, are in serious disagreement with  $C' = 1486 \ \mu\Omega \ \text{cm} \ \text{\AA}$  given in Ref. 26. Unfortunately, it is this last value that was used in the analysis of superconductivity in Nb/Cu superlattices.<sup>4</sup> We believe this value to be incorrect, overestimating the MFP in Nb by a factor of about 4.

Often one does not have a good value for  $\Omega_p$ , but knows the bulk value of e-ph  $h^{(b)}(r)$ , and the e-ph coupling constant  $\lambda_{e-ph}$ . The latter is related to the e-ph scattering rate  $\hbar/\tau$  through the relation<sup>27</sup>



FIG. 2. Dependence of the quantity  $C'_{Nb} = \rho L$  on the residual resistivity  $\rho(0)$ . Calculation for Nb is based on the lifetime-broadened band structure from Ref. 23.

 $\hbar/\tau = 2\pi K_B \lambda_{e-ph} T$ , which is valid above the Debye temperature. Hence, taking  $\rho = h_{id}^{(b)}(295 \text{ K})$  in (11) [strictly speaking, we should use the ideal thermal part  $h_{id}^{(b)}$  rather than the measured  $h^{(b)}$  in this calculation; the difference is often small for metals with low  $\rho(0)$ ], we find

$$C' = \rho L = \left[ \frac{\hbar v_F h_{id}^{(b)}(T)}{2\pi K_B \lambda_{e-ph} T} \right]_{T = 295 \text{ K}}$$
  
= 4.12×10<sup>-7</sup>  $\frac{v_F h_{id}^{(b)}(295 \text{ K})}{\lambda_{e-ph}}$ , (12)

where  $[v_F] = \text{cm/s}$ ,  $[h_{id}^{(b)}] = \mu \Omega \text{ cm}$ , and  $[C'] = \mu \Omega \text{ cm} \text{ Å}$ . For Nb we take  $v_F = 5.6 \times 10^7 \text{ cm/s}$ ,  $^{23} \lambda = 1$ ,  $^6$  and  $h_{id}^{(b)}(295 \text{ K}) = 16.3 \ \mu \Omega \text{ cm}$  [i.e.,  $h_{id}^{(b)} = h^{(b)}/(1 - h^{(b)}/\rho_s)$ , with  $h^{(b)} = 14.5 \ \mu \Omega \text{ cm}$  (Ref. 21) and  $\rho_s = 130 \ \mu \Omega \text{ cm}$ ]. Then we find from (12):  $C'_{\text{Nb}} = 376 \ \mu \Omega \text{ cm} \text{ Å}$ , in good agreement with previously quoted values.

Let us now estimate C' for Al and Cu. In Al,  $v_F = 2 \times 10^8 \text{ cm/s},^{28} h_{id}^{(b)}(295 \text{ K}) \simeq h^{(b)}(295 \text{ K}) = 2.74$   $\mu \Omega \text{ cm},^{21}$  and  $\lambda_{e-\text{ph}} = 0.4;^{29}$  from (12):  $C'_{\text{Al}} = 575 \mu \Omega$ cm Å.

In Cu,  $v_F = 1.57 \times 10^8$  cm/s,<sup>28</sup>  $h^{(b)}(295$  K) = 1.7  $\mu\Omega$  cm,<sup>21</sup> and  $\lambda = 0.13$ :<sup>30</sup> we obtain  $C'_{Cu} = 851$  $\mu\Omega$  cm Å. [We ignored the fact that in Al and Cu the Debye temperature is higher than T = 295 K; the ignored correction amounts to  $\sim (5-7)\%$  in both metals.]

Room-temperature MFP's from our C' values for clean Nb, Al, and Cu are, respectively, 24, 210, and 500 Å. The values of  $\rho_s$  also can be obtained from C', as  $\rho_s = C'/a_n$ . Taking C' given above and  $a_n$  from Kittel,<sup>28</sup> we find  $\rho_s^{\text{Nb}} = 122 \ \mu\Omega \text{ cm}, \ \rho_s^{\text{Al}} = 201 \ \mu\Omega \text{ cm}, \text{ and } \rho_s^{\text{Cu}} = 332 \ \mu\Omega \text{ cm}.$  It is interesting that, while previously quoted, free-electron values were  $\sim 20-30\%$  lower than these band-structure ones, the trend for the three metals is the same, with Nb having the lowest and Cu the highest values. In what follows we use Fig. 2 to find C' for Nb at a given state of disorder  $\rho(0)$ , and use the above constant C' values for Al and Cu.



FIG. 3. Low-temperature MFP's in Nb/Al versus layer thickness. Dashed line at 45° indicates the MFP equal to the layer thickness. Values calculated with and without saturation are shown as indicated.

#### VI. ANALYSIS OF Nb/Al AND Nb/Cu DATA

We have two sets of equations: Equations (4) are simpler, but they do not account for resistivity saturation [i.e., for the decrease of h(T) in more resistive layers]. Equations (9) account for this effect, but they are more involved. It is instructive to use both sets of equations in order to decide whether the use of (9) is justified. Table I contains input parameters  $[d_1, d_2, \rho_m(r), \rho_m(0)]$  for Nb/Al and Nb/Cu, as well as values of resistivity for individual layers calculated through (9). Note that in order to use (9), we need to know  $\rho_s = C'/a_n$ , which in Nb in turn depends on  $\rho_1(0)$ . Hence a self-consistent process has to be adopted: One guesses at  $C'_{Nb}$ , calculates  $\rho_1(0)$ , and checks in Fig. 2 whether the guess was a correct one. It does not take long—one or two iterations are typically



FIG. 4. Low-temperature MFP's in Nb/Cu versus layer thickness  $d_1=d_2=\lambda/2$ .

enough; there is no need to require a perfect agreement between  $\rho_1(0)$  and  $C'_{Nb}$ ; results in terms of  $\rho_1,\rho_2$  are only weakly dependent on  $\rho_s$ . There is no such problem with Al and Cu, where we take C' to be constant. Figures 3 and 4 show values of MFP's  $L_1 = C'_1/\rho_1(0)$  and  $L_2 = C'_2/\rho_2(0)$  for Nb/Al and Nb/Cu calculated with saturation (Table I) as well as without it (not listed in the table). We observe, not a dramatic, but a noticeable difference between the two calculations. Perhaps more significantly, for the thinnest layers simple equations (4) do not have a solution at all (notice the absence of triangular points at the lowest  $\lambda/2$  in Figs. 3 and 4). This clearly indicates that saturation is significant in this short MFP regime—one cannot explain the observed resistivity using constant bulk values of  $h^{(b)}(r)$ .

### VII. DISCUSSION AND CONCLUSIONS

Looking at Figs. 3 and 4, two points can be made. First, contrary to one's naive expectations, the MFP's in individual layers of Nb and Al are not equal to the layer thickness, but are considerably shorter. Out of the four types of layers analyzed in this work (Nb in Nb/Al, Al in Nb/Al, Nb in Nb/Cu, and Cu in Nb/Cu), only Cu in Nb/Cu has MFP which is long enough to be terminated by the interfaces. We suspect that these short MFP's L < d are common in existing metallic superlattices. The assumption that  $L \simeq d$  is probably incorrect in most cases, and should not be made blindly in the analysis of superconducting and tunneling data.

Second, we notice different behavior of L versus d for the two systems studied. In Nb/Al the MFP's in Nb and Al do not differ from each other too much, and show similar behavior with the layer thickness: in both metals the MFP asymptotically tends to a nearly constant value in thicker layers, this value being  $\sim 70$  Å for Nb and  $\sim$  120 Å in Al (Fig. 3). Note that in a single ("bulk") film of Nb, the MFP  $L_{Nb}(0) = C'/\rho(0) = 342/4.64 = 74$  Å (from Table I), limited, no doubt, by the grain size in the Nb film. So it makes perfect sense that in multilayers of Nb/Al the MFP tends to the same value in thicker Nb layers. In "bulk" Al film  $L_{Al}(0) = 575/0.88 = 653$  Å, which is much higher than the corresponding asymptotic value in multilayers. This is rather interesting, for, according to the x-ray work,<sup>17</sup> Nb/Al possesses a fair degree of structural coherency. Perhaps this MFP behavior can be taken as an indication that the degree of perfection in the Al layer (grain size, for one) is strongly influenced by Nb, at least for  $\lambda/2 < 200$  Å.

Behavior in the Nb/Cu system is somewhat different: the MFP in both Nb and Cu increases linearly with  $\lambda/2$ , at least up to  $\lambda/2 \sim 200$  Å (Fig. 4). However, in thinner layers the MFP in Nb is smaller in this system than in Nb/Al, while the MFP in Cu, as was stated above, is limited by the layer thickness and is considerably longer than that in Nb. One can, again, refer to the x-ray evidence,<sup>17</sup> and try to associate this difference in behavior with the apparently better coherency in the Nb/Al system compared to Nb/Cu. One can argue that in coherent systems the MFP's in the two metals are "pulled together," while in less coherent structures the two layers may be independent of each other, and so the MFP's can be rather different, as in Nb/Cu.

In conclusion, our simple methods have been producing sensible results; even the simplified version (4) allows one to see the overall behavior of the MFP in individual layers of a superlattice. Certain parallels with independent x-ray structural analysis can be made. All that one has to know in order to perform the analysis and find the MFP are resistivities at two different temperatures [what we called  $\rho_m(r)$  and  $\rho_m(0)$ ], and layer thicknesses. In closing, we would like to urge investigators of metallic superlattices to present their data in terms of absolute resistivities rather than in terms of easier obtained, but much less useful,

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- <sup>10</sup>We note that Durbin *et al.* (Ref. 8) overestimate all MFP's (and, therefore, the ratio of the MFP to the layer thickness) in Nb/Ta superlattices by about a factor of 3 due to the incorrect starting assumption that L = 70 Å in Nb at room temperature [see Ref. 8(c)]. As we shall see in Sec. V, the correct value is ~25 Å. Nevertheless, at low temperatures the MFP's in Nb/Ta superlattices are still longer than layer thicknesses both from the data of Durbin *et al.* (Ref. 8) and from that of Hertel *et al.* (Ref. 9).

resistivity ratios. The knowledge of the MFP should be useful in the interpretation of various measurements performed on metallic multilayers, in particular superconducting and tunneling ones.

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