

## Coulomb pseudopotential in some disordered Zr-based alloys

Ö. Rapp

*Department of Solid State Physics, The Royal Institute of Technology, S-10044 Stockholm, Sweden*

(Received 18 October 1985; revised manuscript received 27 February 1986)

The Coulomb pseudopotential  $\mu^*$  in McMillan's formula for the superconducting transition temperature  $T_c$  is determined from measurements of the normal-state resistivity and  $T_c$  for a number of disordered Zr-based alloys. The result for  $\mu^*$  is  $0.15 \pm 0.015$ .

### I. INTRODUCTION

In this paper we address the question of whether the Coulomb pseudopotential  $\mu^*$  can be determined for amorphous alloys from measurements of the normal-state resistivity and the superconducting transition temperature  $T_c$ . Such measurements are of interest since there are few alternative methods to determine  $\mu^*$ . In fact, the only two well-known methods, based on the isotope effect<sup>1</sup> and tunneling experiments,<sup>2</sup> respectively, require particular conditions such as a set of isotopes or adequate tunnel junctions with sufficiently strongly coupled superconductors. Furthermore, the problem of the magnitude of  $\mu^*$  in amorphous metals is related to the active field of quantum interference effects in strongly disordered systems. It is generally believed<sup>3,4</sup> that disorder should increase  $\mu^*$ , but the question of whether this can be observed in melt-spun alloys, e.g., in the critical field, remains open.

In Sec. II the present method is described. It is the  $\mu^*$  in McMillan's<sup>1</sup> formula for the superconducting transition temperature  $T_c$  which is determined. The input data for several Zr-based alloys are given and the conditions to which the model can be applied are discussed. There is considerable experimental support for an essential condition in this analysis, i.e., the proportionality between the electron-phonon interaction  $\lambda$  and the temperature derivative at about 270 K of the electrical resistivity. It is possible, but not necessary, for the subsequent analysis to relate this observation to the generalized Ziman model for amorphous metals. In Sec. III the analysis is performed. Since there is some scatter in the input data the result for  $\mu^*$  is tested by repeating this analysis for a successively reduced number of data points. The result for  $\mu^*$  in all cases is within the limits  $0.15 \pm 0.01$ . Variations of input data, due to estimated experimental errors, give about the same variation of the result for  $\mu^*$ . In conclusion, it is found that an average  $\mu^*$  in McMillan's formula for several Zr-based glassy metals can be determined to within 10% to be 0.15.

### II. THE MODEL AND ITS PREREQUISITES

#### A. Method

Assume (i) that  $\lambda$  is proportional to the temperature derivative of the electrical resistivity  $d\rho/dT$  above about the Debye temperature  $\Theta$  for a set of related alloys:

$$\lambda = k d\rho/dT, \quad (1)$$

where  $k$  is a constant and, furthermore, (ii) that  $\mu^*$  is a constant for these alloys. One can then insert Eq. (1) into McMillan's<sup>1</sup> equation for  $T_c$  written in the form

$$\left[ \ln \left( \frac{\Theta}{1.45 T_c} \right) \right]^{-1} = \frac{\lambda}{1.04(1+\lambda)} - \mu^* \frac{1+0.62\lambda}{1.04(1+\lambda)}, \quad (2)$$

and use  $k$  and  $\mu^*$  as parameters to fit experimental data of  $T_c/\Theta$  and  $d\rho/dT$ .

Because of the typical scatter of experimental data, such a fitting procedure will converge satisfactorily only if a large range of  $T_c$  values is covered by the alloys obeying Eq. (1). This situation is rare in crystalline alloys. In noble-metal alloys, for instance, the maximum  $T_c$  is limited by the solubility limit in the fcc phase to about 300 mK. In this case, therefore, a linearized form of Eq. (2) must be used to determine  $k$  and  $\mu^*$ .<sup>5</sup>

In certain cases, Eq. (1) is obeyed also for amorphous alloys with negative  $d\rho/dT$  and  $k$ .<sup>6</sup> Since the measurable  $T_c$  can be varied by more than three orders of magnitude by varying the concentration in many transition-metal-transition-metal glassy alloys these alloy systems seem to offer an interesting possibility to apply the full nonlinear analysis to determine  $k$  and  $\mu^*$ .

#### B. Equation (1) in Zr-based alloys

Experimental values of  $d\rho/dT$  and  $T_c$  are given in Table I for 13 Zr-based glassy alloys. All such samples have been included, for which measurements of  $T_c$  and the temperature coefficient of resistance  $\alpha$  were performed on the same samples in our laboratory. Results for  $\rho$  were read from graphs constructed to average data in the literature. Sources are given in the Table.

The precaution of using only well-controlled samples and of evaluating  $\alpha$  in a consistent way appears to be important. Results in the literature for the temperature dependence of  $\rho$  of disordered metals are sometimes surprisingly different. For instance, different signs for  $da/dc$ , with  $c$  the Zr concentration, have been reported for Zr-Co glasses with  $65 \leq c \leq 80$  at. % (Refs. 10 and 14) and for Zr-Ni glasses with  $70 \leq c \leq 80$  at. %.<sup>15,16</sup> For Nb<sub>0.4</sub>Ni<sub>0.6</sub>,  $\rho$  was found to decrease<sup>17</sup> or increase<sup>18</sup> with increasing temperature in the range up to 300 K.

With  $\Theta = 200$  K and  $\mu^* = 0.15$  according to the result in Sec. III,  $\lambda$  is calculated from the observed  $T_c$  and plotted versus the observed  $d\rho/dT$  in Fig. 1. The choice of  $\mu^*$  does not influence the quality of this fit. Any reasonable  $\mu^*$ , in

TABLE I. Data for Zr-based disordered alloys.

<i>n</i>	Composition <sup>a</sup>	$\rho$ ( $\mu\Omega\text{ cm}$ )	$a^b$ ( $10^{-5}\text{ K}^{-1}$ )	$d\rho/dT$ ( $n\Omega\text{ cm/K}$ )	$T_c^b$ (K)
1	Zr <sub>70</sub> Pd <sub>30</sub>	180 <sup>c</sup>	-11.7	-21.06	2.37
2	Zr <sub>35</sub> Cu <sub>65</sub>	178 <sup>d</sup>	-6.28	-11.18	0.015
3	Zr <sub>79.9</sub> Fe <sub>20.1</sub>	159 <sup>e</sup>	-14.03	-22.30	2.95
4	Zr <sub>80.4</sub> Co <sub>19.6</sub>	162 <sup>f</sup>	-15.12	-24.50	4.04
5	Zr <sub>76.2</sub> Fe <sub>23.8</sub>	162 <sup>e</sup>	-12.48	-20.22	2.03
6	Zr <sub>50</sub> Cu <sub>50</sub>	179 <sup>d</sup>	-9.47	-16.95	0.80
7	Zr <sub>64.6</sub> Co <sub>35.4</sub>	179 <sup>f</sup>	-11.9 <sup>g</sup>	-21.30	2.67
8	Zr <sub>70.7</sub> Co <sub>29.3</sub>	173 <sup>f</sup>	-13.2	-22.84	3.42
9	Zr <sub>70</sub> Ni <sub>30</sub>	169.5 <sup>h</sup>	-12.7	-21.53	2.87
10	Zr <sub>58.6</sub> Co <sub>41.4</sub>	186 <sup>f</sup>	-10.27	-19.10	1.82
11	Zr <sub>66.7</sub> Co <sub>33.3</sub>	177 <sup>f</sup>	-12.75	-22.5	2.82
12	Zr <sub>70</sub> Co <sub>30</sub>	173 <sup>f</sup>	-12.5	-21.63	3.3
13	Zr <sub>65</sub> Cu <sub>35</sub>	167 <sup>d</sup>	-10.7	-17.87	2.0

<sup>a</sup>The samples are from two sources: Nos. 1, 2, 6, 9, 12, and 13 were made by H. S. Chen, the others by Å. Östlund. See Ref. 7.

<sup>b</sup>References 6, 7, and 14.

<sup>c</sup>Reference 8.

<sup>d</sup>References 9–11.

<sup>e</sup>References 10–12.

<sup>f</sup>References 10, 11, 13, and 14.

<sup>g</sup>This is an average value of four measurements. See Ref. 14.

<sup>h</sup>References 10, 11, and 13.

the range, say, 0–0.20, would give a similar scatter with an rms value of about 0.02 in  $\lambda$ . The magnitude of  $\lambda$  and hence of  $k$  depends strongly on the chosen  $\mu^*$ , however, and  $k$  varies from -0.015 to -0.035 K/n $\Omega$  cm in the given range of  $\mu^*$ .

Spin fluctuations could influence Eq. (1). If  $T_c$  for some of the alloys in Table I were depressed by such interactions the value of  $\lambda$  calculated from the observed  $T_c$  by Eq. (2) would be smaller than that determined by the right-hand side of Eq. (1). The observation from Fig. 1 that Zr-Co and Zr-Fe alloys fit Eq. (1) with about the

same scatter as the rest of the data thus suggests that spin fluctuations are not significant in these alloys. For Zr-Co this conclusion is reinforced by results for  $d\rho/dT$  in alloys with low  $T_c$  values.<sup>14</sup> For Zr-Fe the absence of a spin-fluctuation contribution to the resistivity of Zr<sub>72</sub>Fe<sub>28</sub> was recently suggested.<sup>19</sup>

The result in Fig. 1 gives strong empirical support for the validity of Eq. (1) in these alloys. In fact, the results for  $d\rho/dT$  are within  $\pm 5\%$  of a straight line for all data points except one where the deviation is about 10%. This is a small scatter for results for  $d\rho/dT$  of disordered alloys, particularly in view of the differing results for related quantities exemplified above. Moreover, variations in  $\mu^*$  may also affect the scatter in Fig. 1. However, if this were the only cause for the largest deviation from a straight line in the figure,  $\mu^*$  for this sample, Zr<sub>65</sub>Cu<sub>35</sub>, would be decreased by about 0.03. This would seem less likely since the Fermi-surface properties of these alloys are known to be dominated by Zr *d* electrons and a considerable uniformity of electronic properties is expected and indeed often observed. Variations in  $\Theta$  are usually<sup>20–22</sup> within 10% of 200 K and could change the calculated  $\lambda$  by at most about 0.02. Errors in the measurements of  $T_c$  give insignificant changes of  $\lambda$ .

Equation (1) could be explained from the generalized Ziman model for the electrical resistivity of amorphous metals provided certain conditions were fulfilled.<sup>6</sup> For instance, the diameter of the Fermi surface  $2k_F$  must be close to the magnitude of the scattering vector at the first peak in the structure factor. If this model is valid for the present alloys, the expected small variations among the samples in Table I of  $k_F$  and some of the other model parameters would cause  $k$  in Eq. (1) to vary somewhat and thus produce scatter in Fig. 1.

In summary, therefore, the correlation in Fig. 1 is re-

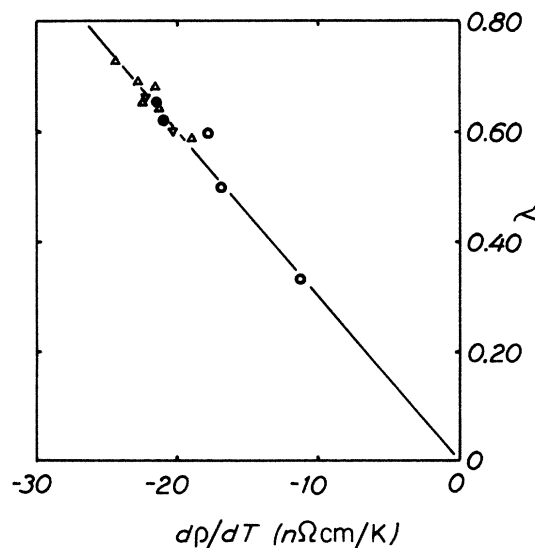


FIG. 1.  $\lambda$  vs  $d\rho/dT$  for the alloys in Table I.  $\Delta$ : Zr-Co,  $\nabla$ : Zr-Fe,  $\circ$ : Zr-Cu,  $\bullet$ : Zr-Pd or Zr-Ni.

markably good. This is a useful empirical fact. One application is the analysis in Sec. III.

### III. RESULTS FOR $\mu^*$

If each alloy system in Table I is separately analyzed one can relax the condition that  $k$  be constant for all alloys and study systematic trends in  $k$  and  $\mu^*$ . Such analyses were found to give less satisfactory results, however, with poorly defined values of  $k$  and  $\mu^*$  due to the scatter of the present input data. To study such trends it would seem that results in each alloy system for alloys with low  $T_c$ 's must be available. In the present case, therefore, only average values of  $k$  and  $\mu^*$  for the alloys of Table I can be determined.

A least-squares fit of all the data of Table I to Eqs. (1) and (2) gives  $\mu^* = 0.140$  and  $k = -0.0292$  K/n $\Omega$  cm. The rms value of the fit to Eq. (1) is 0.02 in  $\lambda$ . This determination of  $\mu^*$  is quite uncertain, however, since variations of  $\mu^*$  within a large range will give almost equally good fits to Eq. (2).

To investigate how reliable such an estimate may be, different subsets of the input data were investigated. The point of view is taken that Eq. (1) is obeyed for some Zr-based alloys. The scatter of the results in Fig. 1 may be due to the beginning of a breakdown of some of the assumptions discussed above or to experimental problems. Therefore, the analysis is repeated with a successively decreasing number of data points. At each step that alloy is removed from the analysis which has the largest deviation from the straight line in Eq. (1). The data in Table I have been arranged in such a way that  $\mu^*(n)$  is the value of  $\mu^*$  obtained from this analysis for the first  $n$  alloys in the table.

Figure 2 illustrates how these procedures affect the quality of the value of  $\mu^*$  determined from Eqs. (1) and (2). For some values of  $n$  the rms error of the fit to Eq. (2) is plotted versus  $\mu^*$ . A  $10^{-3}$  error in  $\{\ln[\Theta/(1.45T_c)]\}^{-1}$  corresponds to a 1% error in  $T_c$  at 4 K and 8% at 15 mK. For each value of  $\mu^*$ , the minimum rms was obtained by varying  $k$  which for  $\mu^*$  in the range 0.11–0.19 required a variation of  $k$  from  $-0.026$  to  $-0.034$  K/n $\Omega$  cm. A conservative statistical estimate would give the results that  $\mu^*$  for the first eight samples in Table I is  $0.15 \pm 0.04$  and for the first six samples is  $0.147 \pm 0.01$ .

Figure 3 shows  $\mu^*(n)$  for the first  $n$  samples in Table I as a function of  $n$ . The rms value of the fit to Eq. (2) is also shown. For all these subsets of the original data the best value of  $\mu^*$  remains within  $0.15 \pm 0.01$ .

Figure 1 suggests that the result may be particularly sensitive to the data for Zr<sub>35</sub>Cu<sub>65</sub> (sample 2 in Table I). Therefore, the analysis was repeated, alloying  $T_c$  for this sample to vary in the range 10–20 mK. This is a much larger temperature range than usually called for by experimental uncertainty but is motivated in the present case by particular conditions.<sup>6</sup> If  $T_c$  for this sample were 20 mK a smaller  $\mu^*$  will result, i.e.,  $\mu^*(13) = 0.13$  and  $\mu^*(6) = 0.135$ . If  $T_c$  were 10 mK the corresponding result would be  $\mu^*(13) = 0.16$  and  $\mu^*(6) = 0.166$ . A possible error of 50% in  $T_c$  of this sample thus gives an uncertainty of about 10% for  $\mu^*$ .

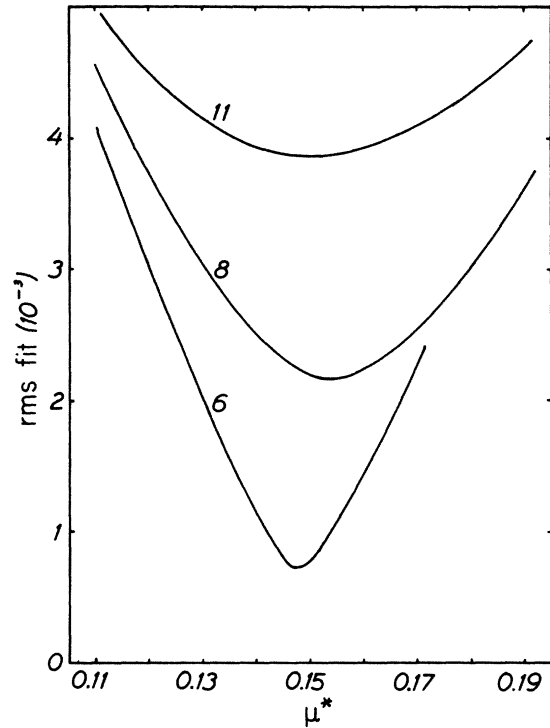


FIG. 2. The rms value of the fit to Eq. (2) as a function of  $\mu^*$ . The calculation is repeated for the 11, 8, and 6 first samples of Table I.

For  $\mu^* = 0.15$ , one finds  $k = -0.0297$  K/n $\Omega$  cm. As mentioned, the rms error of the fit to Eq. (1) is 0.02 in  $\lambda$  for all 13 alloys. In this form, Eq. (1) may provide a useful empirical relation to estimate  $\lambda$  from resistivity data for several disordered Zr-based alloy systems. In the present analysis, the fit to Eq. (1) of course improves with

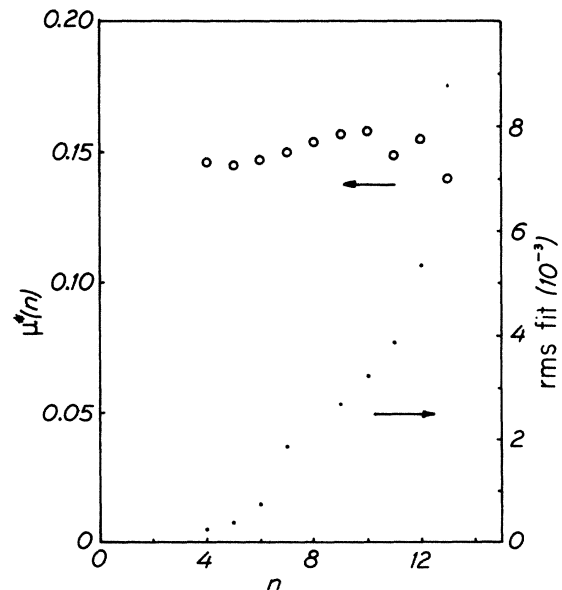


FIG. 3. O:  $\mu^*(n)$  for the  $n$  first samples of Table I as a function of  $n$  (left scale). ●: rms error of the fit to Eq. (2) (right scale).

decreasing  $n$ . The rms value drops rapidly and falls below 0.01 already at  $n=9$ .

A spin-fluctuation contribution to the electron-mass enhancement  $\lambda_{sp}$  has been suggested in several Zr-based amorphous alloys<sup>23</sup> with small values of  $\lambda_{sp}$  for the present alloys. The largest value in Table I would occur for  $Zr_{76.2}Fe_{23.8}$  with  $\lambda_{sp} \approx 0.06$  from Ref. 23 and the present result for  $\mu^*$ . To investigate whether this sample has contributed to a higher value of  $\mu^*$ , the analysis described above was repeated after excluding this sample. The differences from Fig. 3 were insignificant, however, and the result is again  $0.15 \pm 0.01$ .

A possible reason for this difference between Ref. 23 and the present work is the neglect of the late-3d-metal contribution to the orbital part of the magnetic susceptibility in Ref. 23. This approximation would overestimate the exchange enhancement of the Pauli susceptibility and hence also  $\lambda_{sp}$ . This is more serious for Zr-Fe than for the other alloy systems since the merging of the late-transition-metal  $d$  electrons into the Fermi surface is more prominent in this case,<sup>24</sup> and it is more serious in  $Zr_{76}Fe_{24}$  than in  $Zr_{80}Fe_{20}$  due to the larger Fe concentration.

#### IV. CONCLUDING REMARKS

It has been found that an average value of  $\mu^*$  for 13 Zr-based disordered alloys is 0.15. The accuracy is at the level of 10%. The present result is thus somewhat larger than most previous results for  $\mu^*$ , which are around 0.10 for a great number of tunneling experiments<sup>25</sup> and about 0.13 in some other cases.<sup>1,5</sup> This may be due to a similarity to crystalline Zr, for which the absence of a measurable isotope effect would suggest<sup>1</sup>  $\mu^* = 0.17$ . There may also be some disorder induced enhancement of  $\mu^*$ . This possibility is not necessarily in contrast to a similarity to crystalline Zr which has very unusual transport properties for a metallic element.<sup>26</sup>

#### ACKNOWLEDGMENTS

I am grateful to A. Inoue for giving me a copy of Ref. 16 prior to publication and to G. Grimvall for his comments on the manuscript.

- 
- <sup>1</sup>W. L. McMillan, Phys. Rev. **167**, 331 (1968).  
<sup>2</sup>E.g., W. L. McMillan and J. M. Rowell, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), Vol. 1, p. 561.  
<sup>3</sup>S. Maekawa and H. Fukuyama, J. Phys. Soc. Jpn. **51**, 1380 (1981); S. Maekawa, in *Anderson Localization*, Springer Series in Solid State Sciences, Vol. 39, edited by Y. Nagaoka and H. Fukuyama (Springer, Berlin, 1982), p. 103.  
<sup>4</sup>P. W. Anderson, K. A. Müttilab, and T. V. Ramakrishnan, Phys. Rev. B **28**, 117 (1983).  
<sup>5</sup>Ö. Rapp and M. Flodin, Phys. Rev. B **26**, 99 (1982).  
<sup>6</sup>Ö. Rapp, J. Jäckle, and K. Froböse, J. Phys. F **11**, 2359 (1981).  
<sup>7</sup>Ö. Rapp, B. Lindberg, H. S. Chen, and K. V. Rao, J. Less Common Met. **62**, 221 (1978); Ö. Rapp, M. Flodin, Å. Östlund, and H. Fredriksson, Phys. Scr. **25**, 804 (1982); Ö. Rapp, M. Flodin, and L. Hedman, in *Superconductivity in d- and f-Band Metals*, edited by W. Buckel and W. Weber (Kernforschungszentrum, Karlsruhe, 1982), p. 351.  
<sup>8</sup>F. P. Misell and J. E. Keem, Phys. Rev. B **29**, 5207 (1984).  
<sup>9</sup>D. Pavuna, J. Non-Cryst. Solids **61-62**, 1353 (1984).  
<sup>10</sup>E. Babic, R. Ristic, M. Miljak, and M. G. Scott, in *Proceedings of the Fourth International Conference on Rapidly Quenched Metals, Sendai, 1981*, edited by T. Masumoto and K. Suzuki (Institute of Metals, Sendai, 1982), p. 1079.  
<sup>11</sup>Z. Altounian and J. O. Strom-Olsen, Phys. Rev. B **27**, 4149 (1983).  
<sup>12</sup>M. Flodin, Transactions of the Royal Institute of Technology, Report No. FYS 5117, 1984 (unpublished).  
<sup>13</sup>M. G. Karkut and R. R. Hake, Phys. Rev. B **28**, 1396 (1983).  
<sup>14</sup>M. Flodin, L. Hedman, and Ö. Rapp, Phys. Rev. B (to be published).  
<sup>15</sup>Z. Altounian, R. Harris, and J. O. Strom-Olsen, J. Non-Cryst. Solids **61-62**, 1185 (1984).  
<sup>16</sup>A. Inoue, K. Matsuzaki, H. S. Chen, and T. Masumoto, J. Mater. Sci. **21**, 1258 (1986).  
<sup>17</sup>S. R. Nagel, J. Vassiliou, P. M. Horn, and B. C. Giessen, Phys. Rev. B **7**, 462 (1978).  
<sup>18</sup>G. Ilonca, in *Proceedings of the Fifth International Conference on Rapidly Quenched Metals, Würzburg, 1984*, edited by S. Steeb and H. Warlimont (North-Holland, Amsterdam, 1985), p. 1039.  
<sup>19</sup>J. O. Strom-Olsen, Z. Altounian, R. W. Cochrane, and A. B. Kaiser, Phys. Rev. B **31**, 6116 (1985).  
<sup>20</sup>K. Samwer and H. v. Löhneysen, Phys. Rev. B **26**, 107 (1982).  
<sup>21</sup>D. G. Onn, L. G. Wang, and Y. Obi, J. Non-Cryst. Solids **61-62**, 1149 (1984).  
<sup>22</sup>M. Matsuura, U. Mizutani, and K. Fukamichi, in *Proceedings of the Fifth International Conference on Rapidly Quenched Metals, Würzburg, 1984*, edited by S. Steeb and H. Warlimont (North-Holland, Amsterdam, 1985), p. 1019.  
<sup>23</sup>E. Batalla, Z. Altounian, and J. O. Strom-Olsen, Phys. Rev. B **31**, 577 (1985).  
<sup>24</sup>V. L. Moruzzi, P. Oelhafen, A. R. Williams, R. Lapka, H. J. Güntherodt, and J. Kubler, Phys. Rev. B **27**, 2049 (1983).  
<sup>25</sup>P. B. Allen and R. C. Dynes, Phys. Rev. B **12**, 905 (1975).  
<sup>26</sup>E.g., J. Bass, in *Metals: Electronic Transport Phenomena*, Landolt Börnstein New Series, Group 3, Vol. 15, Pt. a, edited by K. H. Hellwege and J. O. Olsen (Springer, Berlin, 1982), p. 91.