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Various methods for determining the critical metallic volume fraction ϕ_c at the metal–insulator transition

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Abstract. The metal–insulator transition in composite Al–Ge films has been extensively studied experimentally. Using the superconducting properties of clusters composed of aluminium grains in the presence of a magnetic field, the critical metallic volume fraction ϕ_c^{SP} has been determined for the case of grain percolation (gp). Because the films investigated are composed of extremely small grains of Al partially coated with amorphous Ge, the critical volume fraction ϕ_c takes on a comparably large value, typically about 56 at.% Al.

The low-temperature electrical conductivity data are examined using a number of criteria for ϕ_c . Most of these criteria yield values ϕ_c to within 1% of ϕ_c^{SP} . However, one widely used criterion involving the fitting of the low-temperature conductivity data to a $T^{1/2}$ dependence, as suggested by the electron–electron interaction theory, was found to yield a significantly smaller value for ϕ_c . Methods based on room-temperature data are also evaluated. These criteria yield ϕ_c values that are within $\pm 1\%$ of ϕ_c^{SP} .

1. Introduction

Over the last 20 years, there has been extensive research on the metal–insulator transition (MIT) in crystalline, amorphous and granular materials. The MIT occurs at the critical volume fraction ϕ_c , which separates metallic films from insulating films. Metallic films in the normal state have finite electrical conductivities at absolute zero in temperature, while insulating films have zero conductivities at absolute zero. This paper concentrates on the granular Al–Ge system [1]. Such films consist of Al grains embedded in an amorphous Ge matrix doped with Al. If the matrix has an infinite resistivity and if the Al grains have a macroscopic size ($> 100 \text{ \AA}$), then the metallic content ϕ_c at the metal–insulator transition can, in principle, be easily determined from the classical percolation conductivity equations [2].

However, quantum-mechanical tunnelling between metallic grains (as well as dopant atoms in amorphous and crystalline materials) complicates the conduction process. The grains appear to be electrically ‘connected’ to one another even when their separation distance is of the order of tens of Ångströms. In addition, the ‘insulating’ matrix has a small but finite electrical conductivity, even at liquid-helium temperatures, which provides a significant alternative conduction path in samples close enough to ϕ_c [3]. Therefore, most systems exhibit a smooth change in the electrical resistivity or conductivity at finite temperatures, as the metal content is decreased through the MIT [3]. This behaviour contrasts

with the sharp change in the conductivity or divergence in the resistivity, predicted by percolation theory [2]. Consequently, in most systems that should exhibit a percolation type of behaviour, it is not obvious how to identify uniquely the critical metallic volume fraction ϕ_c ; that is, how to discriminate between metallic and insulating samples at finite temperatures.

To some extent, the MIT granular systems should be comparable to the MIT in homogeneous amorphous films and to the MIT in doped crystalline semiconductors; correspondingly, the symbol ϕ in granular systems (the symbol x instead of ϕ is used by many investigators) is analogous to n , the effective carrier concentration in doped semiconductor systems. Some of the methods discussed in this paper are found to be applicable not only to a granular metal embedded in an insulating high-resistivity matrix but also to the MIT in amorphous semiconductors alloyed with transition metals and to the MIT in doped crystalline semiconductors.

Scaling theory is used to discuss the experimental results [4]. It implies that the conductivity goes continuously to zero as the MIT is approached from above [4], in contrast to the Mott conductivity hypothesis [5, 6]. Unfortunately, our experimental data for the Al-Ge system, close to the MIT, do not show sufficient detail to differentiate between both predictions [4-6].

We first described the experimental methods for preparing the Al-Ge films and extracting ϕ_c^{EP} from the superconductivity data. We then examine the various criteria used to define ϕ_c from the low-temperature measurements and conclude with an examination of high- (room-) temperature criteria for the MIT.

2. Film preparation and experimental measuring techniques

Composite 2000 Å Al-Ge films were fabricated by co-evaporation of Al and Ge, using two electron guns, onto glass slices at room temperature [7]. In order to obtain the amorphous form of the Ge matrix yielding ϕ_c values close to 55% Al, it was important to maintain the substrates near room temperature [1, 8]. Evaporations were performed in a vacuum of 3×10^{-6} mmHg at rates of 5 to 10 Å s⁻¹. Twenty-one small glass slices having dimensions of 2.5 mm × 25 mm were glued onto one ordinary glass microscope slide, which was positioned above and at nearly the same distance from both sources. After the evaporation, the Al volume fractions (ϕ) on neighbouring sample slices differ slightly, the film closest to the Ge target having the highest insulator content. A span in the Al content of about 20% was found to occur between the two extreme slices. By varying the relative evaporation rates, several series were obtained covering different Al concentration regions.

The volume fraction ϕ was determined using an EDAX (energy-dispersive analysis of x-rays) probe attached to an electron microscope. The ϕ values are given in atomic per cent Al in the following sections. The relative Al concentrations from film to film within a series are known to about 1% from EDAX measurements, while the absolute values determined from both EDAX and the relative evaporation rates are known only to $\pm 5\%$. Fortunately, none of the methods described in this paper require knowing the absolute value of the Al concentration, with the exception of using the general effective medium (GEM) equation.

Transmission electron microscope studies have shown that the germanium matrix is amorphous [9]. Owing to the doping of the germanium by the aluminium, the germanium is not an ideal insulator [3]. For the insulating films having ϕ less than ϕ_c , the Al grains are smaller than 20 Å, and in fact they are so small that the grains cannot be seen directly in the electron microscope [10]. However, these amorphous metallic grains in the form of

clusters can and do exhibit enhanced superconducting transition temperatures of 1.6 to 2 K. Above the MIT, the diameters of the Al crystalline grains rapidly increase to 100–200 Å [10]. For all except the very highest ϕ samples, the Al grains are coated or partially coated with an amorphous Ge layer.

Resistance measurements between room temperature and 4.2 K were performed in a 'dip-stick' probe that was slowly lowered into a commercial liquid-helium storage dewar [3]. Between 4.2 and 0.45 K, resistance measurements were performed in a ^3He adsorption refrigerator equipped with a small superconducting magnet capable of producing a 3.5 T field perpendicular to the films [11]. A calibrated Speer 470 Ω , 0.5 W carbon resistor, which showed negligible magnetic-field dependences even at 0.45 K, was used as a thermometer [12]. The film resistance was measured using either a HP 3456 microvoltmeter or a Keithley 617 electrometer in its manual ranging mode, in order to control and limit the measuring current. As the samples could not be immersed directly into the liquid He, very small currents had to be used to avoid Joule heating of the films, even at the cost of degrading the accuracy of the resistance measurements. Therefore, in the ^3He adsorption refrigerator, resistances of the most insulating films were measured to an accuracy of only $\pm 5\%$. For the metallic films the accuracy is considerably better.

The normal-state conductivity data are used in the methods to be discussed. Since the Al in these Al-Ge films went superconducting below 2 K, it was important that above T_c all superconducting fluctuation effects including the Maki-Thompson [13, 14] and Aslamazov-Larkin [15] conductivities, be completely quenched, and that below T_c all superconductivity effects be eliminated. This was accomplished by applying a 3.5 T field perpendicular to the films. The granular Al-Ge system is unique in that such a small field can completely quench the superconductivity. In most other small grain systems, much larger fields are required.

3. Results and discussion

3.1. The extraction of ϕ_c^{gp} from superconducting magnetoresistance measurements

The basic idea of the present section is to utilize the unique magnetic-field dependences of the resistivities of all the films in their superconducting state to determine the critical volume fraction of aluminium, ϕ_c^{gp} , at the percolation threshold of the grains, where gp refers to grain percolation.

An earlier study showed that the small-diameter Al clusters and grains go superconducting below 2 K [11]. In the metallic films, the clusters and small grains (≈ 20 Å) form continuously connected backbones. In order to drive these backbones entirely normal, very large fields in excess of 3 T are required. Elsewhere [16, 17] a physical explanation is given of why the critical field is enhanced in very small-diameter filaments by over two orders of magnitude from the bulk value of 0.01 T for Al. For example, in the series No 9 films, all films having Al content of 51.2% Al or greater exhibited zero resistivities up to large fields of 1.5 T.

In contrast, all the films having 50.1% Al content or less in the No 9 series exhibited finite measurable resistances in moderately small magnetic fields of 0.05 T or greater. We interpret such behaviour as Josephson junction coupling between disconnected clusters and/or disconnected filaments that form the current-carrying backbone. The methods described in this paper do not rely on knowing the normal-superconducting boundary at ϕ_c^{sc} . Only the percolation threshold ϕ_c^{gp} is considered. Note that we have not employed the special precautions suggested by Barber and Dynes in measuring the transitions [18].

The critical Al content ϕ_c^{SP} is defined as midway between the last sample to show Josephson junction coupling and the first sample to show zero resistivity in fields above 1 T at 0.5 K [3, 11]. Thus, this magnetic-field behaviour of the superconducting Al-Ge films provides a diagnostic criterion for separating the percolating films from the non-percolating films, or equivalently for separating the metallic films from the insulating films. The above criterion yields a sharp boundary; it permits us to estimate ϕ_c^{SP} to within $\pm 0.5\%$ Al. The experimental results for ϕ_c^{SP} for several series of Al-Ge samples, as well as the associated room-temperature and 5 K conductivity data for the individual series, are summarized in table 1. The reader is referred to figure 1 and 2 in [11] for additional details.

Table 1. Experimental values for the critical metal volume fraction ϕ_c^{SP} , obtained from superconductivity measurements, for five different Al-Ge series. The table also includes the room-temperature conductivity and resistivity values at ϕ_c^{SP} as well as the 5 K conductivity values.

Al-Ge series number	ϕ_c^{SP} (% Al)	σ_c (300 K) $((\Omega \text{ cm})^{-1})$	ρ_c (300 K) ($\Omega \text{ cm}$)	σ_c (5 K) $((\Omega \text{ cm})^{-1})$
3	59.4	93.4	0.0107	36.6
4	55.9	73.6	0.0136	35.8
8	56.7	82.3	0.0122	41.3
9	50.7	83.0	0.0112	52.5
12	54.9	64.8	0.0154	30.1

The knowledge of reliable experimental values for $\phi_c^{\text{SP}} = \phi_c$ motivated us to make a comparative examination of various methods of determining ϕ_c . These methods are discussed in the following sections.

3.2. Methods of extracting ϕ_c from low-temperature conductivity data

3.2.1. Theoretical discussion. The scaling theory predicts that the zero-temperature conductivity $\sigma(0)$ goes continuously to zero as a function of composition as the MIT is approached from the metallic side [4] and that there should be no Mott minimum metallic conductivity. Therefore, according to this theory, the MIT takes place at the metal volume fraction $\phi = \phi_c$ where the zero-temperature conductivity vanishes.

The concept of a minimum metallic conductivity requires that the zero-temperature conductivity should vanish discontinuously at a point ϕ_c where the minimum metallic conductivity is given by $\sigma_{\text{Mott}} = 0.03e^2/\hbar a$ [5, 6]. The quantity σ_{Mott} takes on the appropriate value of $240 (\Omega \text{ cm})^{-1}$ provided that the spatial extension of the Al states, a , is taken as the relevant length and estimated to be 3 Å. However, if the mean grain size, 20 Å, is regarded as the relevant parameter, one obtains $\sigma_{\text{Mott}} \simeq 36 (\Omega \text{ cm})^{-1}$. The latter estimation presumes that the dopant Al atoms in the Ge matrix can be neglected with respect to the MIT. As the prefactor 0.03 is known only to within a factor of 2, this condition $\sigma(T=0) = \sigma_{\text{Mott}}$ can never give a precise value of ϕ_c .

The concept of a finite minimum conductivity is not widely accepted today. However, as can be seen in table 1, the values of $\sigma_c(300 \text{ K})$ and $\sigma_c(5 \text{ K})$, related to grain percolation as obtained in the previous section, are remarkably constant with average values of about 80 and 40 $(\Omega \text{ cm})^{-1}$. The latter value agrees well within the above estimate based on the grain size as the relevant length scale. Moreover, it should be noted that the temperature dependence of the samples considered is very weak and flat. Extrapolations according to

$$\sigma(T) = \sigma(0) + CT^{1/2}$$

yield $\sigma(0)$ values between 54 and 92 ($\Omega \text{ cm}$)⁻¹ for the two metallic samples (the 51.2% and 52.5% Al samples in the No 9 series) closest to the MIT. These observations can be considered as an argument in favour of a finite minimum metallic conductivity.

The majority of MIT investigators start from the scaling theory and determine their critical volume fraction ϕ_c by extrapolating low-temperature electrical conductivity data, usually taken well below 1 K, to absolute zero in temperature. Most investigators use the expression

$$\sigma(T) = \sigma(0) + CT^y \quad (1)$$

with $y = 1.2$ and C being a positive constant. Then, $\sigma(0)$ is plotted against ϕ ; and the value of ϕ where $\sigma(0) = 0$ is taken to be ϕ_c . Equation (1) with $y = 1/2$ has its origin from the electron-electron interaction theory in three dimensions [19]. Weak localization contributions to the conductivity are usually neglected below 1 K. According to the electron-electron interaction theory [19], the constant C can be either positive or negative depending upon the strength of the electron screening constant F [20]. A majority of systems investigated exhibit a positive C , i.e. the conductivity slowly decreases with decreasing temperature. Unfortunately, the theory mentioned is a perturbation theory and at this stage it is not known whether or not equation (1) with $y = 1/2$ is still valid very close to the MIT, where strong interaction effects must be considered [20]. At present, there is no conductivity theory near the MIT in any dimension that considers strong electron-electron interaction and strong localization effects on an equal footing [20]. Therefore equation (1) with $y = 1/2$ is probably valid only for 3D films having metallic contents considerably greater than ϕ_c .

Another problem concerning the applicability of equation (1) with $y = 1/2$ results from not knowing the exact distribution of the dopant metal atoms in crystalline and amorphous materials or the exact morphology of the metal grains in granular systems. Al chains and clusters, composed from 20 Å grains, form a continuous backbone through the film just on the metallic side of the MIT. The chains and clusters that form the conducting backbone could, in some sections, show 1D effects rather than 3D effects. If weak 1D interaction effects are dominant, then the conductivity would follow equation (1) with $y = -1/2$ instead of 1/2 [20]. The situation becomes still more complicated when contributions to the conductivity from the matrix have to be taken into account [3, 21]. We assume the Al-doped Ge matrix to exhibit activated behaviour close to ϕ_c . Therefore, in granular systems, it is not clear what exponent should be used in equation (1).

Several investigators studying crystalline, amorphous or granular systems have plotted their conductivity data using an exponent y smaller than 1/2. A convincing derivation for $y = 1/3$ was given by Maliepaard *et al* [22]. These workers started from the relation

$$\sigma(T) = Ae^2/\hbar\xi + Be^2/\hbar L_{in}(T).$$

They presumed the constant-conductivity part to be small in comparison to the T -dependent part owing to the divergence of the correlation length ξ as the MIT is approached. Moreover, they assumed the interaction length $L_{in}(T)$ to be roughly equal to the thermal diffusion length $L_T = (D\hbar/k_B T)^{1/2}$ and to be the only relevant length scale [23]. Eliminating the diffusion constant by means of the Einstein relation, $\sigma = e^2 DN(E_F)$, they obtained $y = 1/3$ [22]. Previously, the exponent 1/3 was obtained by Ovadyahu starting from other assumptions [24, 25].

However, it is not clear whether the dominant length in granular systems is the thermal diffusion length $L_T(T)$. In general, the inelastic length is given by $L_{in}(T) = [D\tau_{in}(T)]^{1/2}$,

where $\tau_{\text{in}}(T)$ is the inelastic scattering time. In granular Al-Ge composites near the MIT, the system contains thin Al filaments or wires. According to Wind *et al* [26] the dominant time in thin aluminium wires is the Nyquist time, which has a $\tau \propto T^{2/3}$ dependence [27]. We estimated that the Nyquist time is even smaller [27] (stronger scattering) than the thermal diffusion time $\tau_T = \hbar/k_B T$ in thin Al wires and thus is the dominant scattering time. Therefore, provided the constant-conductivity part is small in comparison to the T -dependent part for σ , the $\tau \propto T^{-1/3}$ dependence implies that

$$\sigma(T) = \sigma(0) + \bar{C}T^{2/9} \quad (2)$$

and thus, $x \simeq 0.22$.

In granular systems, it is not clear which value of the exponent should be used in equation (1). Thus, in the next section, we adjust not only the parameters $\sigma(0)$ and C , but also y . To the best of our knowledge, the only investigators who have previously taken this approach are Möbius [28], Audouard *et al* [29], Raychaudhuri [30] and Rajeev and Raychaudhuri [31]. In the amorphous SiCr system, Möbius observed that the exponent y took on the low value of 0.19 ± 0.03 for films exhibiting negative $d\sigma/dT$ values as $T \rightarrow 0$ [28].

For an insulating film, one normally assumes that the conductivity can be described by the general hopping law

$$\sigma(T) = \sigma_0 \exp[-(T_0/T)^x] \quad (3)$$

where T_0 is a characteristic temperature and x an exponent. As the characteristic temperature T_0 is believed to scale inversely with the localization length ξ with some critical exponent and since ξ diverges at the MIT, T_0 should approach zero at the MIT [32]. Commonly observed values of x are 1/4 for Mott variable-range hopping in 3D and 1/2 for variable-range hopping in a Coulomb gap [32]. It should be noted that equation (3) can be physically meaningful only if $T < T_0$.

3.2.2. Analytical approaches. The applicability of equation (1) can be checked by plotting the logarithmic derivative $S_{\text{ln}} = d \ln \sigma / d \ln T$ versus T for each film [33, 34]. Using equation (1) one gets

$$S_{\text{ln}} = d \ln \sigma / d \ln T = -yCT^y / [\sigma(0) + CT^y]. \quad (4)$$

This equation predicts that, for metallic films above the MIT, where $\sigma(0)$ is finite, S_{ln} should tend to zero as the temperature approaches absolute zero, no matter what the sign of C might be and no matter which value y has, provided only that $y > 0$. If the transition is continuous, i.e. $\sigma(0) = 0$, equation (4) yields $S_{\text{ln}} = y$ for a film exactly at the MIT, independently of the measuring temperature. Moreover, equation (4) predicts the vanishing of S_{ln} to occur at increasingly lower temperatures, viz. for $T < [\sigma(0)/C]^{1/y}$, as the MIT is approached from the metallic side.

In contrast, for an insulating film where T_0 is a characteristic temperature and x an exponent, we obtain using equation (3)

$$S_{\text{ln}} = d \ln \sigma / d \ln T = -x(T_0/T)^x. \quad (5)$$

S_{ln} is always positive and diverges with decreasing temperature. The divergence clearly occurs at increasingly lower temperatures as the metal content approaches the MIT from below since $T_0 \rightarrow 0$ as $\phi \rightarrow \phi_c$.

Therefore, three different kinds of behaviour should be observed with continuity of the MIT presumed: (i) Metallic films not close to the MIT will always exhibit a S_{in} that tends to zero with decreasing temperature. (ii) Insulating films, sufficiently far from the MIT, should show a S_{in} that increases to infinity with decreasing temperature. (iii) However, very close to ϕ_c , S_{in} should be nearly temperature-independent for insulating films with T_0 values smaller than the lowest experimentally accessible temperature, as well as for metallic films with $[\sigma(0)/C]^{1/y} \ll T$. If the minimum metallic conductivity hypothesis is assumed, then S_{in} should be temperature dependent right up to the transition.

On the insulating side, very close to the MIT, equation (3) probably has to be modified. Often a decrease of x from $\frac{1}{2}$ down to $\frac{1}{4}$ is observed as the MIT is approached. Moreover, the exponential equation (3) has surely to be modified if the lowest measuring temperature exceeds T_0 . Nevertheless, on the metallic side, we used a simple, more or less empirical, form with the correct asymptotic behaviour to characterize the samples. In the same sense, it might be helpful to utilize equation (3) on the activated side for an empirical description without theoretical reasoning. The exponential term in equation (3) can be approximated in a series expansion as

$$\sigma(T) \simeq \sigma_0[1 - (T_0/T)^x + \dots] \simeq \sigma_0 - C^*T^{-x}. \quad (6)$$

Comparing equation (6), which describes insulating films, with equation (1), which describes metallic films, suggests that the exponent in the temperature power law will change sign as well as the prefactor when crossing the MIT. Thus, our third (new) criterion is the simultaneous change of signs of y and C from positive to negative values.

Until now, the applicability of equation (1) characterizing the conductivity of metallic films was checked. Now, we assume that equation (1) is valid and we describe the methods used to determine the free adjustable parameters: $\sigma(0)$, C and y . The vanishing of $\sigma(0)$, as well as the change of the signs of y and C , could be considered as indications of the MIT.

The three parameters appearing in equation (1) can be determined by fitting equation (1) to all the low-temperature data using a suitable computer program. Alternatively, equation (1) can be manipulated to eliminate $\sigma(0)$ and C , using three conductivity data points ($\sigma_1, \sigma_2, \sigma_3$) taken at three temperatures (T_1, T_2 and T_3). This gives an equality

$$(\sigma_2 - \sigma_3)/(\sigma_1 - \sigma_2) = (T_2^y - T_3^y)/(T_1^y - T_2^y) \quad (7)$$

that y must satisfy. When the value of y has been estimated, both C and $\sigma(0)$ can easily be calculated. Moreover, a derivative approach, similar to the method suggested by Zabrodskii and Zinov'eva can be used [35]. If the data are sufficiently smooth to obtain accurate numerical derivatives, then one can obtain y from

$$y = 1 + \{\ln[\Delta\sigma(T_1)/\Delta T_1] - \ln[\Delta\sigma(T_2)/\Delta T_2]\}/(\ln T_1 - \ln T_2) \quad (8)$$

which is derived from equation (1). For the samples studied, equations (7) and (8) gave values of y that agree to $\pm 5\%$.

Summarizing, the following low-temperature criteria are available for determining the critical Al content ϕ_c :

- (i) The logarithmic derivative $d \ln \sigma / d \ln T$ changing its qualitative behaviour as $T \rightarrow 0$.
- (ii) Vanishing of the parameter $\sigma(0)$, obtained from fitting equation (1), related to metallic films, using the free adjustable parameter y .
- (iii) Vanishing of the parameter $\sigma(0)$, obtained from fitting equation (1) with y fixed to $1/2$.
- (iv) Change of sign of the exponent parameter y and the prefactor parameter C obtained from the free parameter fit of equation (1).
- (v) Vanishing of the characteristic temperature parameter T_0 in the hopping expression, equation (3), describing insulating films.

3.2.3. *Data analysis.* The above criteria can now be tested using the experimental low-temperature normal conductivity data for the Al-Ge No 9 series. Corresponding overviews of $\rho(T, B = 3.5 \text{ T})$ and of $\sigma(T, B = 3.5 \text{ T})$ are given in figures 1(a) and (b). Note that it is impossible to determine the MIT from inspection of these two figures. In the following procedures, we neglect the very small positive magnetoresistance of 1%.

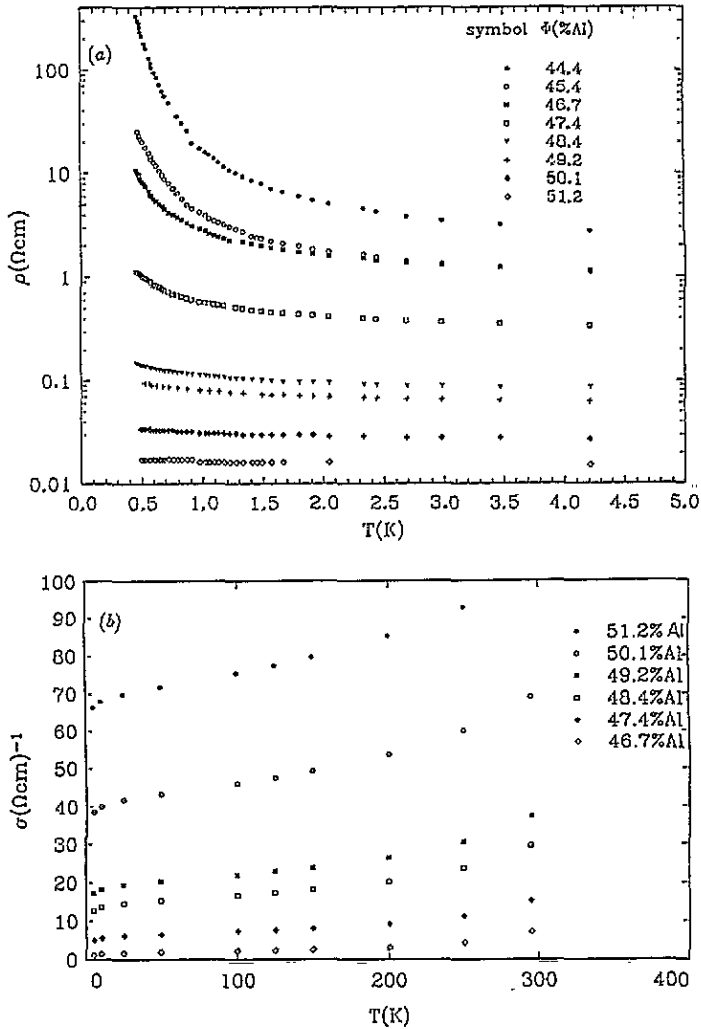


Figure 1. (a) Low-temperature resistivity data in the normal state versus temperature. All superconductivity effects were quenched with the application of a 3.5 T magnetic field. It is impossible to deduce the location of the MIT by simple inspection of these curves. (b) High-temperature conductivity data versus temperature. These data were used to deduce ϕ_c using the 'knee' method described in the paper and illustrated in figure 7.

The first criterion (i) is illustrated in figure 2, which shows a plot of $S_{\text{In}} = d \ln \sigma / d \ln T$ against T for the No 9 series near ϕ_c . The 52.5%, 53.5% and 54.3% Al films, which exhibit a decrease in S_{In} with decreasing temperature, should be metallic. In contrast, the 50.1%

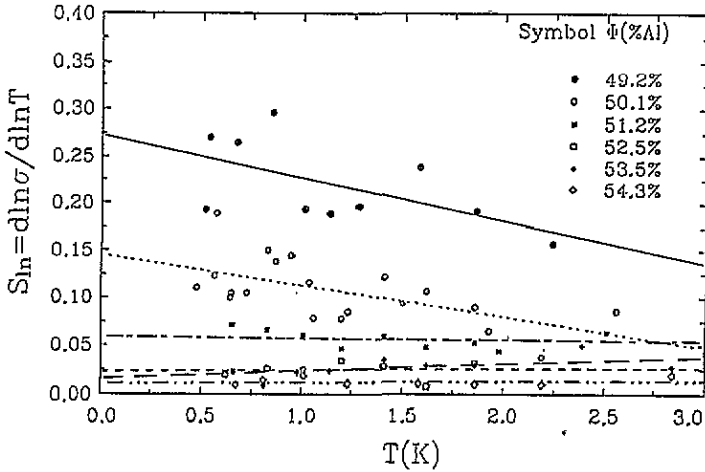


Figure 2. $S_{in} = d \ln \sigma / d \ln T$ versus T for the No 9 series. Films with 52.5%, 53.5% and 54.3% Al show a decrease in S_{in} with decreasing temperature. Films with 50.1% and 49.2% Al show an increase in S_{in} with decreasing temperature. The film with 51.2% Al shows a nearly temperature-independent behaviour of S_{in} and is probably located at the MIT.

and 49.2% Al films, which show an increase of S_{in} with decreasing temperature, should be insulating. The virtually temperature-independent behaviour of the 51.2% Al film indicates that this sample is extremely close to the MIT. A ϕ_c of 51.2% Al is in good agreement with the ϕ_c^{sp} value of 50.7% Al. Note that, owing to the experimental errors in the data, the limiting value $S_{in} = 0$ at $T = 0$ K for the metallic samples is not observed. Owing to the very small values of T_0 of the 48.4%, 49.2% and 50.1% Al samples, the lowest measuring temperature of 0.5 K is not low enough for making visible the sharp divergence of S_{in} as $T \rightarrow 0$.

Now we turn to criterion (ii), i.e. to the consideration of the behaviour of $\sigma(0)$ using the adjustable parameter y . The parameter y was determined using both equation (7) and/or (8). For the data just above ϕ_c , which had the lowest experimental error, the results for y were found to agree within 5%. Small values for the parameter y , ranging between 0.09 to 0.36, were obtained for the different metallic films. The values of $\sigma(0)$ are plotted against ϕ in figure 3. A linear extrapolation to $\sigma(0) = 0$ gives a ϕ_c of about 51.7% Al, within 1% of the ϕ_c^{sp} value of 50.7% Al.

A large number of investigators have utilized method (iii); that is, they used equation (1) with $y = 1/2$ to determine ϕ_c . We followed the same procedure. Figure 4 shows that the plot of $\sigma(0)$ versus ϕ is non-linear, with an exponent clearly larger than 1. Furthermore, $\sigma(0)$ becomes negative for $\phi < 47.8\%$ Al, when extrapolated to zero according to the $(\phi - \phi_c)^2$ power law. Thus, this standard method deviates significantly from the $\phi_c^{sp} = 50.7\%$ Al result. The possible overestimation of the $\sigma(0)$ values for $\text{Ge}_{1-x}\text{Au}_x$ from extrapolations within $y = 1/2$ were previously noted in [36]. For Si:P, it is shown [33, 34, 37] that extrapolations with y fixed at $1/2$ are erroneous very close to the MIT.

Another estimate of ϕ_c can be obtained from method (iv) in which y and C , appearing in equation (1), change sign at the MIT. Both y and C changed their signs between the 51.2% and 52.5% Al films. Taking the average of these two values gives $\phi_c \simeq 51.8\%$ Al, close to ϕ_c^{sp} . Note that these changes of sign come from an unbiased determination of y , C and $\sigma(0)$ from the data, using either equation (7) or (8).

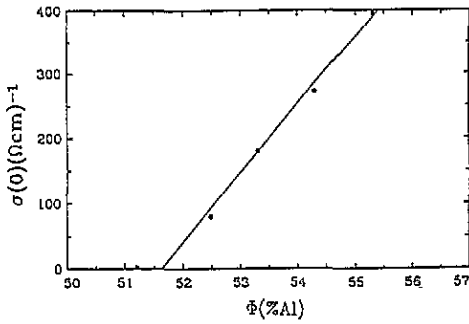


Figure 3. The fitting parameter $\sigma(0)$ for the metallic films in the No 9 series, as a function of Al content, ϕ . The values of $\sigma(0)$ were obtained by fitting equation (1) to the conductivity data in a field of 3.5 T, using the temperature power-law exponent γ as one of the three fitting parameters. The full line is the linear extrapolation used to obtain ϕ_c .

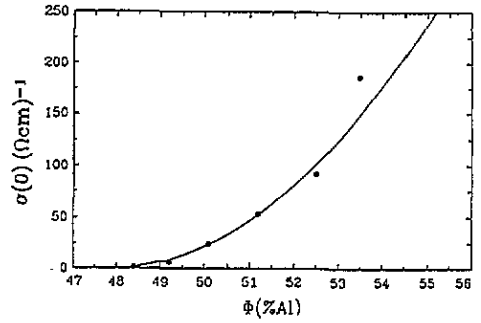


Figure 4. The parameter $\sigma(0)$, obtained by a fit to the conductivity data of the No 9 series using the expression $\sigma(T) \approx \sigma(0) + CT^{1/2}$, plotted against ϕ . The full curve shows the quadratic expression $\sigma(0) = 45800(\phi - 0.478)^2$, from which $\phi_c = 47.8\%$ Al. This value should be compared to $\phi_c^{BP} = 50.7\%$ Al.

Method (v) is based on measurements on the insulating side of the transition. According to equation (3), the resistivity should obey the general hopping relation

$$\rho(T) = \rho_0 \exp(T_0/T)^x. \quad (9)$$

The parameters T_0 , ρ_0 and x are functions of ϕ . Method (v) consists in determining the point $\phi = \phi_c$ where T_0 vanishes. To our knowledge, the method was applied first to amorphous $\text{Si}_{1-x}\text{Cr}_x$ and crystalline Ge [37–39]. The parameters ρ_0 , T_0 and x for series No 9 were obtained from the normal resistivity data of the insulating films, using the fitting procedure suggested by Zabrodskii and Zinov'eva [35]. A plot of T_0 versus ϕ is shown in figure 5. A linear extrapolation of T_0 to zero in figure 5 yields $\phi_c \simeq 49.2\%$, fairly close to ϕ_c^{BP} . Only the four T_0 values greater than 1/2 K were used in the extrapolation.

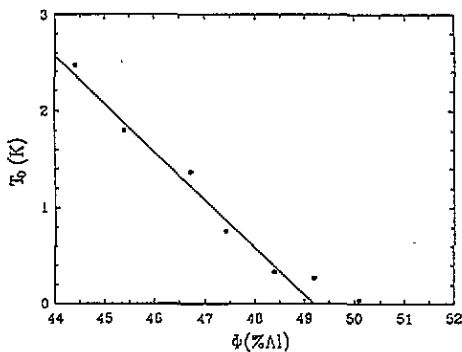


Figure 5. The characteristic temperature T_0 , obtained by fitting the insulating data of the No 9 series to equation (9), against ϕ . The full line is the linear extrapolation used to find ϕ_c , using the four highest data points only. Recall that $\phi_c^{BP} = 50.7\%$ Al.

3.3. Methods for determining ϕ_c from room-temperature measurements

As it is always easier and more convenient to make measurements at room temperature, various methods of extracting ϕ_c from this type of data are of particular interest.

3.3.1. The percolation and GEM equations. The percolation equations for the measured (m) electrical conductivity and resistivity are, for $\phi \geq \phi_c$ and σ_{lo} equal to zero (the insulating component is a perfect insulator) [2, 21],

$$\sigma_m = \sigma_{hi}(1 - f/f_c)^t = \sigma_{hi}[(\phi - \phi_c)/(1 - \phi_c)]^t \quad (10)$$

while for $\phi \leq \phi_c$ and ρ_{lo} equal to zero (the metallic component is a perfect conductor),

$$\rho_m = \rho_{hi}(1 - \phi/\phi_c)^s. \quad (11)$$

In equation (10), σ_m is the electrical conductivity of the composite system (medium in the case of a continuum percolation system), $\sigma_{hi} = 1/\rho_{lo}$ is the conductivity of the conducting component, $f = 1 - \phi$ is the volume fraction of the perfect insulator (or fraction of perfectly insulating sites or bonds), $f_c = 1 - \phi_c$, and t is one of the conductivity exponents. In equation (11), ρ_m is the resistive (finite conductivity) component, ϕ is the volume fraction of the perfectly conducting component (or the fraction of perfectly conducting sites or bonds), ϕ_c is the volume fraction where the perfectly conducting component becomes continuous, and s is the second conductivity (or dielectric) exponent.

From these equations, Efros and Shklovskii [21] and also Straley [40] derived the following expression as a definition of the crossover region

$$|\Delta\phi| = |\Delta f| = (\sigma_{lo}/\sigma_{hi})^{1/(t+s)} = (\rho_{lo}/\rho_{hi})^{1/(t+s)}. \quad (12)$$

The crossover region $|\Delta\phi|$ is the region centred on both sides of ϕ_c where the two percolation equations are not valid owing to the finite conductivity of the insulating component above ϕ_c and to the non-perfect conductivity of the metallic component below ϕ_c . The above authors estimated the conductivity at the critical volume fraction ϕ_c to be [21, 40]

$$\sigma(\phi_c) \simeq (\sigma_{lo})^{t/(t+s)}(\sigma_{hi})^{s/(t+s)}. \quad (13)$$

The GEM (general effective medium) equation written in terms of the electrical conductivity and the volume fraction f of the low-conductivity component σ_{lo} is [41, 42]

$$\frac{f[(\sigma_{lo})^{1/t} - (\sigma_m)^{1/t}]}{(\sigma_{lo})^{1/t} + [f_c/(1 - f_c)](\sigma_m)^{1/t}} + \frac{(1 - f)[(\sigma_{hi})^{1/t} - (\sigma_m)^{1/t}]}{(\sigma_{hi})^{1/t} + [f_c/(1 - f_c)](\sigma_m)^{1/t}} = 0 \quad (14)$$

where all the symbols have been previously defined. For the case when $\sigma_{lo} = 0$, the GEM equation reduces to equation (10), and when $\sigma_{hi} = \infty$ or equivalently $\rho_{lo} = 0$, it reduces to equation (11) [41, 42]. The usual procedure is to fit equation (14) to the σ_m versus ϕ data in order to determine the parameters ϕ_c , t , σ_{lo} and σ_{hi} . The last two parameters are often known already from experimental measurements. The room-temperature resistivity as a function of volume fraction ϕ for series Nos 0, 4, 11 and 12, but not No 9, of Al-Ge samples is shown in figure 6. Data from the No 9 series were not included in the GEM fit as this series had a ϕ_c^{gp} about 10% lower than the ϕ_c s of the other series, resulting in a ρ versus ϕ curve that was considerably lower than those of the other series. The fit using the GEM equation to the data gives $\phi_c = 55.9\%$ Al, $t = 3.46$, $\rho(\text{Ge}) = \rho_{hi} = 76 \Omega \text{ cm}$ and

$\rho(\text{Al}) = \rho_{t0} = 7.7 \mu\Omega \text{ cm}$ [3]. The GEM equation using these parameters is shown by the full curve in figure 6. The broken curves are obtained by fitting the data on the metallic side to the percolation equation, equation (10), and on the insulating side to the percolation equation, equation (11), excluding from the fit all the data points lying in the crossover region defined by equation (12). The parameters for this fit are a common value for $\phi_c = 56.0\%$ Al, $t = 3.35$, $s = 2.92$, $\rho(\text{Ge}) = 61 \Omega \text{ cm}$ and $\rho(\text{Al}) = 7.5 \mu\Omega \text{ cm}$ [3]. As the ϕ_c values determined by the superconductivity magnetoresistance (MR) measurements on the Nos 4 and 12 series are 55.9% Al and 54.9% Al respectively [3], the agreement between these values and those obtained from the GEM and percolating equations is remarkable. However, as this may be coincidental, further systems must be investigated using these two methods. The values of t and s observed are higher than the accepted theoretical universal values of 2.0 and 0.75 [43, 44], and possible reasons for this are advanced in [42]. Note that the data from series No 0 come from [7]. Samples from the series No 11 are all insulating, i.e. not superconducting. It is conceivable that a second MIT transition associated with the amorphous doped Ge matrix may occur. Had such a second transition been present, a smaller 'inverted S' curve would be present in figure 6 on the insulating side of ϕ_c .

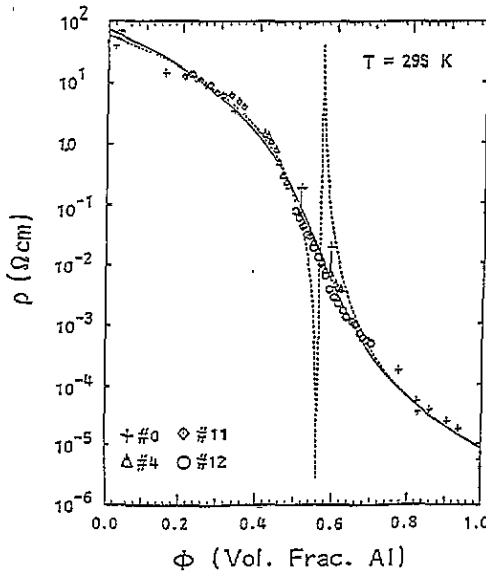


Figure 6. Experimental and theoretical resistivities versus Al volume fraction ϕ at 295 K. The experimental points are the Nos 0, 4, 11 and 12 series. The full curve is a plot of the GEM equation (equation (14)) and the broken curves are plots of the percolation equations (equation (10) and (11)). The parameters used in these plots are given in the text. The daggers indicate the experimental points closest to ϕ_c that are excluded in the fits to equation (10) and (11). The fitting parameter ϕ_c took on the value of 56.0% Al, to be compared with $\phi_c^{\text{SP}} = 55.9\%$ Al for series No 4 and $\phi_c^{\text{SP}} = 54.9\%$ Al for series No 12.

3.3.2. Identification of $\sigma(\phi_c)$ and ϕ_c from $d\sigma/dT$ versus σ data. When a plot of $d\sigma/dT$ versus σ evaluated at room temperature was made for a series of amorphous $\text{Si}_{1-x}\text{Cr}_x$ samples that spanned the MIT [45], it was observed that: (i) for the insulating films, the slope $d\sigma/dT$ increased as the MIT was approached from below and (ii) for the metallic films, the slope was roughly constant in value. The intercept of the extrapolations of the two lines through these two regions can be used to determine $\sigma(\phi)_c$ and hence ϕ_c from the

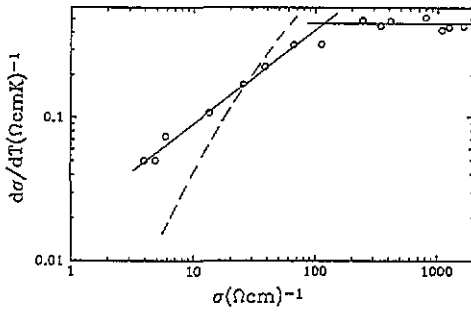


Figure 7. Slopes $d\sigma/dT$ of series No 9, evaluated at room temperature, as a function of the room-temperature conductivity $\sigma(300\text{ K})$. According to Möbius [45] the intersection of the extrapolation, shown by the two full lines, gives the value of the conductivity at ϕ_c . The intersection occurs at $\sigma(\phi_c) \simeq 125\ (\Omega\text{ cm})^{-1}$, which has a corresponding ϕ_c of 51.5% Al, to be compared with $\phi_c^{\text{EP}} = 50.7\%$ Al. The broken curve is obtained from the Thouless conductivity expression discussed in the text.

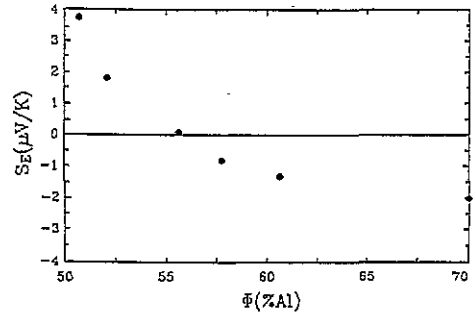


Figure 8. Room-temperature thermoelectric power, S_E , as a function of aluminium metal content ϕ for the No 4 series in the region where S_E changes sign. The ϕ_c^{EP} for the No 4 series is 55.9% Al.

σ versus ϕ curve. The same type of analysis will now be performed on the granular Al-Ge data.

An examination of figure 7, where $d\sigma/dT$ is plotted against σ , reveals a ‘knee’ type pattern. From figure 7, the slope $d\sigma/dT$ increases from 0.05 to 0.5 $(\Omega\text{ cm K})^{-1}$ as the Al content is increased from 44.4% to 50.2% Al. We note from figure 7 that the slope $d\sigma/dT$ has a constant value close to 0.5 $(\Omega\text{ cm K})^{-1}$ for $\phi \geq 51.2\%$ Al. The intercept between the extrapolations of lines passing through these two regions has a σ of about 125 $(\Omega\text{ cm})^{-1}$. A 125 $(\Omega\text{ cm})^{-1}$ sample in series No 9 would have an Al volume content of about 51.5% Al; again this value is reasonably close to what we believe to be the true value of 50.7% Al. This method certainly must be tested in other systems.

Possible explanations for the behaviour of $d\sigma/dT$ versus σ on both the insulating and the metallic sides of the MIT are suggested in [24]. On the insulating side, the increasing values of $d\sigma/dT$ as the MIT is approached from below can be explained using the Thouless [46] conductivity expression,

$$\sigma(T) \simeq e^2 N(E_F) \xi^2 / \tau_{\text{in}}(T)$$

and the Schmid [47] electron-phonon scattering time formula,

$$\tau_{\text{in}} \propto T^{-3}.$$

On the metallic side, the scaling expression,

$$\sigma(T) \simeq Ae^2/\hbar\xi + Be^2/\hbar L_{\text{in}}(T)$$

can be used to explain the observed results [24]. The term $Ae^2/\hbar\xi$ gives the conductivity dependence on metal content ϕ since $\xi = \xi_0/(\phi - \phi_c)^{\nu}$, with $\nu \simeq 1$, and the second term $Be^2/\hbar L_{\text{in}}(T)$ gives the constant magnitude for $d\sigma/dT$. Note that $L_{\text{in}}(T) = [D\tau_{\text{in}}(T)]^{1/2}$,

where again the Schmid electron-phonon scattering time may be used for τ_{in} [47]. The broken curve in figure 7 is calculated using the Thouless expression and the Schmid scattering time. The diffusion constant $D \simeq 2 \text{ cm}^2 \text{ s}^{-1}$ was determined from the $N(E_F)$ procedure suggested in [30] and [31]. Similar values for D were also obtained from critical field measurements.

3.3.3. Thermoelectric power criterion for determining ϕ_c . This possible method of determining ϕ_c experimentally would appear to be limited to systems where the insulating and metallic components have thermoelectric powers or Seebeck coefficients, S , of opposite signs. For aluminium, $S_M = -1.7 \mu\text{V K}^{-1}$, where the minus sign arises from the fact that the carriers are electrons. For the amorphous aluminium-doped germanium, S_I would appear to be in the region of $+1000 \mu\text{V K}^{-1}$ [48]. The positive sign of S_I arises if low-lying acceptor levels are created when Al (valence 3) dopes Ge (valence 4), which gives rise to holes. Therefore, for granular Al-Ge, there must always be an Al concentration at which the thermoelectric power changes sign. Figure 8 shows the thermoelectric power versus ϕ near the MIT, for the No 4 series at room temperature [48]. The zero crossing occurs at 55.3% Al, very close to the experimental ϕ_c^{BP} of 55.9% for this series No 4 (table 1).

In order to check if there is any theoretical justification for the zero crossing of the thermoelectric power at the MIT, the Bergman-Levy expression for the effective thermoelectric power S_E is examined [49]. For ϕ close to ϕ_c , the Bergman-Levy expression for S_E of the composite system can be simplified to

$$S_E \simeq S_M + S_I(K_E\sigma_I/K_I\sigma_E) \quad (15)$$

where S_M and S_I are the thermoelectric powers for the pure metal component and pure insulating component and K_E (σ_E) and K_I (σ_I) are the thermal (electrical) conductivities of the effective (E) composite system and pure insulating system respectively. Based on the estimates for $K(\phi_c)$ and $\sigma(\phi_c)$, using percolation theory given by Efros and Shklovskii [21] as well as Straley [40], Levy and Bergman write for S_E at ϕ_c [50]

$$S_E(\phi_c) \simeq S_M + S_I(K_I\sigma_M/K_M\sigma_I)^{-s/(t+s)} \quad (16)$$

where s and t are the critical exponents appearing in the conductivity percolation equations (11) and (10). Therefore, if $S_E = 0$ at ϕ_c and if S_I and S_M have opposite signs, then $-S_M/S_I = (K_I\sigma_M/K_M\sigma_I)^{-s/(t+s)}$. The authors can see no fundamental reason why this identity should always be satisfied.

We note that experimentally $-S_M/S_I$ ranges from between 1/250 and 1/1500 [48]. From the electrical conductivity data, we know that $\sigma_M/\sigma_I \simeq 10^7$. We estimate $K_I/K_M \simeq 1/25$. It has also been observed earlier from the percolation fits to the electrical conductivity data that $t \simeq s$. Using these values, we obtain that $-S_M/S_I \simeq 1/600$, within a factor of 2 of the experimental values obtained from the fitting procedure [48]. Thus, the theory predicts that the zero crossing should occur slightly below ϕ_c . Perhaps it is fortuitous that the zero crossing of the thermoelectric power in Al-Ge occurred so close to the experimental ϕ_c^{BP} . Certainly this method must be tested on other systems. A serious discrepancy exists between the theory [49, 50] and experiment [48] if the accepted universal values of $t \simeq 2.0$ and $s \simeq 0.76$ are used [43, 44], for in this case we obtain $-S_M/S_I = 1/35$.

3.4. Additional methods

Tunnelling at low temperatures [51, 52] and dielectric measurements [53, 54] are powerful techniques for identifying ϕ_c . These methods are outside the scope of this paper and have not been tested on the Al-Ge system by us.

4. Conclusion

The conclusion of this paper is that, while the method of obtaining ϕ_c using equation (1) with $y = 1/2$ is not a good method for granular and perhaps other systems, there exist a number of methods that appear to give consistent results for ϕ_c to within $\pm 1\%$. These methods need to be further tested and explored, with particular emphasis on more fundamental understanding about them.

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