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The Fermi surface of random-metallic alloys and the oscillatory magnetic coupling across alloy spacers

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Abstract. Calculated Fermi surfaces of $\text{Cu}_{1-x}\text{Ni}_x$ binary alloys are used to interpret measurements of the periods of oscillations in the magnetic coupling across $\text{Cu}_{1-x}\text{Ni}_x$ alloy spacers. The lack of observation of damping of such oscillations is shown to be the consequence of the mean free paths on the Fermi surface being long compared with the oscillation periods. It is argued that oscillatory magnetic coupling is an important new technique for probing the Fermi surfaces of random binary alloys.

1. Introduction

The defining property of the metallic state is the Fermi surface and therefore its measurement has been at the centre of interest in metal physics. The existence of the Fermi surface in metallic systems, as identified by the relation of its features with observed properties of the system, is the most compelling evidence for the validity of the Fermi liquid picture for electrons in metals.

Experimental techniques related to the de Haas–van Alphen oscillations [1, 2] have been proven very powerful tools in the ‘fermiology’ and most of the pure-metal Fermi surfaces have been studied [1] using these techniques more than two decades ago. Unfortunately, the application of these techniques is restricted to the case of pure metals or random binary alloys with very small concentration (less than 1% [3]) due to the scattering (introduced by disorder) of the electrons in the quasiparticle states on the Fermi surface. Indeed, the sufficient condition for the applicability of these techniques [1, 2] is $\omega_c \tau \gg 1$, where ω_c is the cyclotron frequency (eB/m_e for a given magnetic field \mathbf{B}). If the average time between collisions is small, enormous magnetic fields are required for the study of the Fermi surface of the material. An interpretation of the above condition is that the energy differences of the Landau levels in the presence of a magnetic field must be larger than the uncertainty (due to disorder) in the energy of these levels. Equivalently, the time required for a closed orbit in the presence of a magnetic field must be shorter than the lifetime of the quasiparticle states at the Fermi surface. For random binary alloys, even for very large magnetic fields and relatively small concentrations, this condition is far from being satisfied.

An alternative probe which in principle could be applied to the study of the Fermi surfaces of binary alloys for any concentration is the angular correlation of annihilation radiation (ACAR) experimental technique [4]. It does not require the electronic states to

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have long lifetimes and can be used to study Fermi surfaces in so far as they are well defined. The Fermi surface of a random binary alloy is thought to be well defined when the interband energy distances are much larger than the energy uncertainty \hbar/τ introduced by disorder, or equivalently when the wavevector uncertainty δk is much smaller than the linear sizes of the Fermi surface, i.e. the size of the bellies, necks or pockets that might exist on the Fermi surface. The method up to now has had to rely on theoretical calculations of the Fermi surface for the comparison of its measurements [5–7]. For example in reference [5] the neck of the Fermi surface of $\text{Cu}_{1-x}\text{Ni}_x$ calculated with KKR-CPA electronic structure method was compared with ACAR measurements [8, 9]. These theoretical calculations usually combine an electronic structure method (usually *ab initio*) with the CPA ‘mean-field’ approach which is the most accurate ‘single-site’ approximation to the random-binary-alloy problem [10, 11].

During the last decade a surprising new probe appeared, to compete with ACAR for measuring the alloy Fermi surfaces, namely the oscillatory magnetic coupling in metallic multilayers. The observation that two ferromagnetic layers are coupled antiferromagnetically through a metallic spacer [12] and that this coupling oscillates [13], switching between ferromagnetic and antiferromagnetic as the spacer thickness increases, is one of the most fascinating discoveries in solid state physics during the last decade. The oscillatory behaviour was very soon related to the Fermi surface of the bulk spacer material [14, 15]. In particular every extremal vector \mathbf{Q} on the bulk spacer Fermi surface parallel to the direction of growth has been proven to be the wavevector of an oscillation in the interaction of two magnetic layers across the spacer. In the case of a pure-metallic spacer, all of the theoretical approaches [14–16] for obtaining the interaction energy $\delta E(L)$ of the ferromagnetic layers as a function of the spacer thickness L lead to an asymptotic expression of the form

$$\delta E(L) = -\frac{1}{L^2} \sum_{\nu} A_{\nu} \cos(\mathbf{Q}_{\nu} L + \phi_{\nu}) \quad (1)$$

at spacer thicknesses L large compared with the monolayer size. In the above formula the summation is over all of the extremal vectors \mathbf{Q}_{ν} on the bulk spacer Fermi surface, and A_{ν} and ϕ_{ν} are the amplitude and the phase of each of the contributing oscillatory terms. It should be mentioned that the $1/L^2$ law might be modified to an $1/L^{3/2}$ or $1/L$ law in the cases of extremal vectors which are constant in one of or both of the in-plane directions respectively [17]. In the case of a simple metallic spacer, such as Cu, the theory is in very good quantitative agreement with experiment [14, 15]. The discreteness of the spacer thickness, which is always an integer number of monolayers, results in an aliasing effect [14, 15, 18], i.e. the fact that among all of the equivalent (due to periodicity) extremal vectors, the smallest ones correspond to the measured periods.

In the case of binary alloy spacers, which is the topic of the present work, we have recently shown [19] that a similar formula to equation (1) is valid, with an additional exponential damping factor multiplying each oscillatory term in equation (1). In reference [19], employing a simple tight-binding description for the electronic structure and a simple phenomenological model for the disorder, the interaction energy $\delta E(L)$ in the case of a disordered spacer was proven to be

$$\delta E(L) = -\frac{1}{L^2} \sum_{\nu} \bar{A}_{\nu} \cos(\bar{\mathbf{Q}}_{\nu} L + \bar{\phi}_{\nu}) \exp\left(-\frac{L}{\Lambda_{\nu}}\right). \quad (2)$$

In the above equation the index ν counts the extremal vectors $\bar{\mathbf{Q}}_{\nu}$ on the properly defined alloy Fermi surface, \bar{A}_{ν} and $\bar{\phi}_{\nu}$ are again the amplitudes and the phases of each term and, finally, the Λ_{ν} are the characteristic lengths for the damping of each oscillatory term. In

reference [19] an expression of the form

$$\frac{1}{\Lambda_\nu} = \frac{1}{\lambda_\perp(\mathbf{k}_\nu^+)} - \frac{1}{\lambda_\perp(\mathbf{k}_\nu^-)} \quad (3)$$

was found, where \perp indicates projection of the mean free path $\lambda(\mathbf{k})$ in the growth direction and \mathbf{k}_ν^+ and \mathbf{k}_ν^- are the end-points of the ν th extremal vector on the Fermi surface parallel to the direction of growth. The meaning of equations (2) and (3) is that like in the case of pure metal, the periods of oscillations are a direct measure of the extremal vectors of the bulk spacer Fermi surface and in addition the question of exponential damping is answered by the very definition of the Fermi surface itself. In other words, if the Fermi surface of the alloy is well defined, then Λ_ν is much larger than the oscillation period $2\pi/Q_\nu$ and we should not expect any significant damping within the range of the first few oscillation periods. Of course equations (1) and (2) are valid for spacer thicknesses L which are large compared with the monolayer thickness, but for relatively large Λ_ν there is a range of values of L for which the asymptotic expression (2) is valid while the oscillations are not yet damped. It is interesting that, according to equations (1) and (2), the periods of oscillations are not affected by the properties of ferromagnetic layers and are related only to the topology of the Fermi surface of the spacer. This is proven experimentally to be true at least for Cu spacers for Fe and Co ferromagnetic layers (page 96 of [15]).

Unfortunately, not all of the periods predicted by the extremal-vector analysis have been observed even in the case of pure-metal spacers. In particular the small-size periods are not observed and these are thought to be suppressed by the interlayer surface roughness [14, 15, 20]. In addition, calculations of the oscillation amplitudes [21–24] indicate that there must be a difference of several orders of magnitude in the amplitudes of the oscillations originated by different extremal vectors. Such an analysis has only been performed for pure-metallic spacers so far, but there is little doubt that this is the case for alloy spacers as well. Another drawback is the discreteness of the spacer thickness which also restricts the accuracy in the measured periods to the order of magnitude of the spacer monolayer size. Finally, we would like to mention two factors that might affect the agreement between the experimental periods and those predicted from the bulk random-alloy Fermi surface. The first is related to the randomness of the alloy in the experimental samples. Indeed, it might be possible for concentration waves to exist in the alloy layer especially close to the FM layers. This work suggests that these concentration waves, if they exist, are not strong enough to destroy the agreement between the periods predicted by the random-alloy Fermi surface study and the experimental ones. The second is the small spacer thickness in the experimental samples in connection with the definition of the spacer Fermi surface. Apparently, the theoretical models which relate the oscillation periods to the bulk spacer Fermi surface make use of the approximation that the spacer layer thickness tends to infinity. Fortunately, first-principles calculations for Cu spacers for the (100) direction [21], where the periods predicted by total-energy calculations are compared with the ones predicted by the Fermi surface analysis, indicate that the asymptotic limit is correct. On the other hand, the separations of adjacent AF peaks, in experiments [25–27] where more than two such peaks were observed, is more or less constant. Thus, there is relative convergence to the asymptotic limit for spacer thicknesses in the range studied in these experiments.

Nevertheless, in reference [19] the agreement between the measured oscillation periods for $\text{Cu}_{1-x}\text{Ni}_x$ alloy spacers and the ones theoretically predicted from the extremal-vector analysis of the alloy Fermi surface is remarkable. On the other hand the lack of observation of damping in these oscillations is related in reference [19] to the large size of the calculated

mean free paths on the Fermi surface.

In this work the results of [19] are summarized and viewed from the alloy Fermi surface point of view. In section 2 the concept of the alloy Fermi surface is discussed. The agreement between the calculated Fermi surface and the one measured experimentally by means of oscillatory magnetic coupling in $\text{Cu}_{1-x}\text{Ni}_x$ is demonstrated in section 3. Furthermore, in section 3, we suggest that the lack of observation of oscillation damping strongly indicates that the alloy Fermi surface is very well defined at the region of the extremal vectors involved. Measurements of oscillatory magnetic coupling provide a method that is alternative and complementary to positron annihilation techniques (ACAR) for the study of binary alloys' Fermi surfaces. Thus positron annihilation work does not now have only theoretical calculations of alloy Fermi surfaces for suitable comparison.

2. The alloy Fermi surface

Evidently, in the case of disordered systems such as random binary alloys, the Bloch wavevector \mathbf{k} is not a good quantum number. On the other hand, for systems with a periodic underlying lattice, the ensemble of configurations may be invariant under lattice translations. Thus, on average the k -space representation is still valid in spite of the fact that Bloch states are not eigenstates of the system. Of course, the k -space language is useful only if the Bloch states have long enough lifetimes $\tau(\mathbf{k})$ and the energy uncertainty $\hbar/\tau(\mathbf{k})$ is small compared with the interband energy separations, or equivalently the inverse mean free paths of the Bloch states are small compared with the interband distances in k -space. Indeed, it has been proven that this is the typical case for random binary alloys, so the k -space description of the electronic states, on average, is valid and the Fermi surface of many of these systems is well defined. As we discuss later, even small necks and pockets of the pure-host-metal Fermi surface survive in alloying even for large concentrations of the solute and these features are identified in ACAR [8, 9] as well as oscillatory magnetic coupling experiments [25–27]. Since for most of the binary alloys the Fermi surface is well defined across the whole range of concentrations, it is of great interest to see how the shape and connectivity of the Fermi surface change with concentration. Such changes which might be discontinuous have been studied theoretically [28] and referred to as electronic topological transitions.

The basic idea that has been employed for the theoretical study of random binary alloys is to consider the alloy A_{1-x}B_x as a system with its constituents A and B so well mixed that each lattice site has the same probabilities x and $1 - x$ of being occupied by atoms of type A and B respectively. In that sense, all sites are equivalent and a 'mean-field' approximation may be helpful. Of course the ideal solution of the problem would be to average the interesting quantities over all of the configurations of the system. Since that is impossible, approximations for this procedure need to be found. The best 'single-site' approximation to the problem at hand is the coherent potential approximation [10, 11], in which the actual crystal disordered potential is replaced by an effective, energy-dependent, complex, but periodic coherent potential. Thus the average Green's function \bar{G} is diagonal in k -space.

A convenient quantity which makes the above ideas more transparent is the Bloch spectral function (BSF) $A_B(E, \mathbf{k})$. By definition it is the number of states per energy and \mathbf{k} . In the case of a pure periodic system the BSF either as a function of \mathbf{k} or as a function of E is a sum of δ -functions located at these \mathbf{k} (or E) where the dispersion relation $E_{\mathbf{k},\nu} = E$

is satisfied. Thus

$$A_B(E, \mathbf{k}) = \sum_{\nu} \delta(E - E_{\mathbf{k},\nu}) = -\frac{1}{\pi} \text{Im} \left\{ \sum_{\nu} \lim_{\epsilon \rightarrow 0} \frac{1}{E - E_{\mathbf{k},\nu} + z i \epsilon} \right\} = -\frac{1}{\pi} \text{Im} \{G(E; \mathbf{k}, \nu)\} \quad (4)$$

where $G(E; \mathbf{k}, \nu)$ is the Green's function (the resolvent of the periodic crystal Hamiltonian) in k -space. Of course the BSF for constant energy and equal to the Fermi energy along a particular k -direction is a sum of delta functions which are located at the intersection points of the particular direction and the Fermi surface.

In the binary alloy case we should consider the configurationally averaged Green's function

$$\bar{G}(E; \mathbf{k}, \nu) = \frac{1}{E - E_{\mathbf{k},\nu} + \Sigma(E; \mathbf{k}, \nu)} \quad (5)$$

where the disorder effect is included in the complex self-energy $\Sigma(E; \mathbf{k}, \nu)$. Self-energy is a function of E and \mathbf{k} , but if we assume that its imaginary part is small, we can regard it as constant in a neighbourhood of E_1 and \mathbf{k}_1 such that

$$E_1 - E_{\mathbf{k}_1,\nu} + \text{Re}\{\Sigma(E_1; \mathbf{k}_1, \nu)\} = 0$$

for a given ν . Then, writing the $A_B(E, \mathbf{k})$ in terms of \bar{G} it is easy to show that the BSF (for constant $\mathbf{k} = \mathbf{k}_1$) are Lorentzian-like functions of E centred at E_1 . Similarly, for constant $E = E_1$ the BSF are also Lorentzian-like functions of \mathbf{k} centred at \mathbf{k}_1 . Thus, the effect of alloying is to shorten and broaden the δ -like peaks of the BSF, transforming them into Lorentzian-like functions. This is correct in the limit of small disorder, i.e. a small imaginary part of $\Sigma(E; \mathbf{k}, \nu)$, but as we have already mentioned, most of the random binary alloys are close to that limit.

Just as for a pure-metal case, in the one-electron picture these Bloch-like states can be fully occupied up to some energy which could be regarded as the Fermi energy E_F of the system. Unlike in the pure-metal case, however, these electron states at this energy have an energy uncertainty even for zero temperature owing to their finite lifetimes. Nevertheless, a suitable definition of the alloy Fermi surface can still be given in terms of $A_B(E; \mathbf{k})$, as the loci of the peaks of the Lorentzian-like BSF for constant energy $E = E_F$ in k -space. Naturally, this is only valid if the BSF fits well to a Lorentzian sum, which is correct in the limit of small disorder.

The most compelling evidence for the validity of the above picture is the relation of the features of the alloy Fermi surface with observed properties of the material, and ACAR data on binary alloys, as well as the oscillatory magnetic coupling across alloy spacers, are probably the most striking examples of such cases.

3. The $\text{Cu}_{1-x}\text{Ni}_x$ alloys' Fermi surfaces

In this section the calculated Fermi surfaces for $\text{Cu}_{1-x}\text{Ni}_x$ ($x < 0.4$) binary alloys are compared with measurements from oscillatory magnetic coupling experiments. The non-relativistic KKR-CPA electronic structure method [10, 11, 29] was employed for the calculation of the BSF with constant energy $E = E_F$. For the concentrations encountered, the Fermi surfaces of $\text{Cu}_{1-x}\text{Ni}_x$ are topologically the same as that of pure Cu and the extremal vectors for (111), (100) and (110) directions are analogous to the ones also referred to in [14] in the case of pure Cu. These extremal vectors are shown in figure 1.

For the (110) direction, for which four different periods are theoretically predicted, a single one is observed [27]. The neck diameter ($Q_{(110)}^{(2)}$ in figure 1(c)) fits well to that period

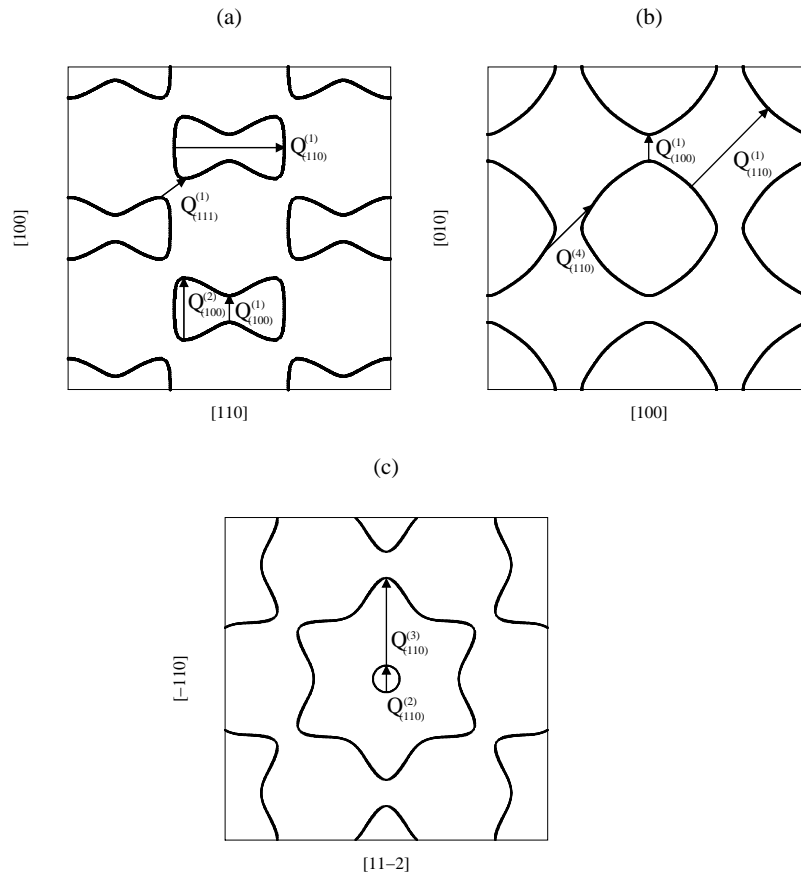


Figure 1. The extremal vectors on the Fermi surface of pure Cu plotted in the repeated-zone scheme. (a), (b) and (c) correspond to three different planes (cuts) in the k -space: on the plane which is perpendicular to the $[1\bar{1}0]$ direction at distance $\Delta k = 0$ to the Γ point (a), on the plane perpendicular to the $[001]$ direction at $\Delta k = 0$ (b) and on the plane perpendicular to the $[111]$ direction and at $\Delta k = \sqrt{3}/2$ (c).

size and is identified as the extremal vector corresponding to this oscillation [14, 27, 19]. In figure 2 the calculated neck diameter as well as the one measured using ACAR [8, 9] and the one predicted from the oscillatory magnetic coupling period for the (110) direction [27] are plotted as functions of the Ni concentration. The agreement between the three different sets of data is striking. The small amplitude size might explain why the other three predicted periods originating from the extremal vectors $Q_{(110)}^{(1)}$, $Q_{(110)}^{(3)}$ and $Q_{(110)}^{(4)}$ have not been observed, while their small oscillation period sizes lead one to suspect that the interlayer surface roughness might also be a reason. Interlayer surface roughness obviously affects the small oscillation periods more drastically than the large ones [14, 15, 20].

In figure 3(a) another large calculated oscillation period for the (111) direction is shown as a function of the concentration and compared with experimental values from references [25, 26]. Again, there is very good agreement between theory and experiment concerning both the size and concentration dependence of the period. The extremal vector corresponding to that oscillation is $Q_{(111)}^{(1)}$ shown in figure 1(a), also being in the area of the neck.

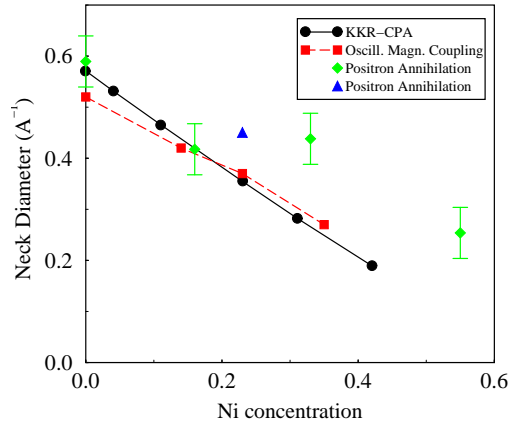


Figure 2. The evolution of the diameter of the neck in the Fermi surface of $\text{Cu}_{1-x}\text{Ni}_x$ binary alloys with Ni concentration x , compared with oscillatory magnetic coupling and positron annihilation experiments. The squares and dashed line correspond to the oscillatory magnetic coupling experiment of reference [27], while the diamonds with the error bars and the triangles correspond to positron annihilation (ACAR) experiments from references [8] and [9] respectively.

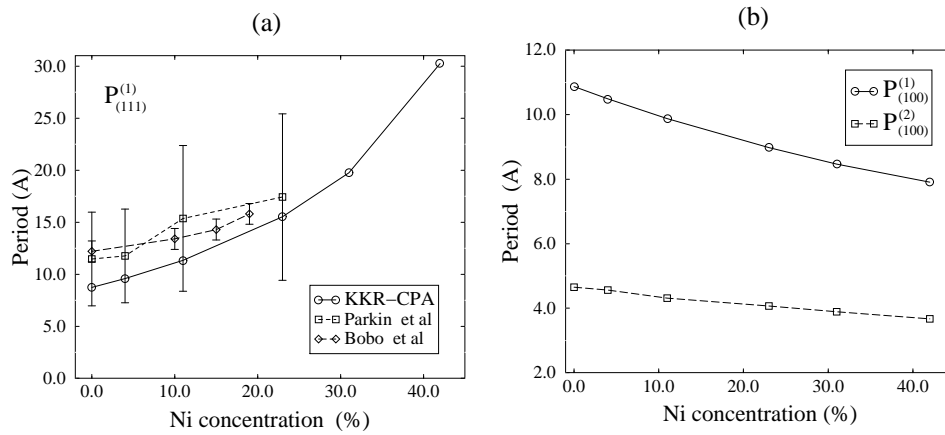


Figure 3. The evolution of the calculated periods $P_{(klm)}^{(i)} = 2\pi[Q_{(klm)}^{(i)}]^{-1}$ with Ni concentration for the (111) (a) and (100) (b) directions of growth. In figure 1 the corresponding extremal vectors $Q_{(klm)}^{(i)}$ are shown. In (a) the experimental values from Parkin *et al* [25] and Bobo *et al* [26] are also included. The extremal vector $Q_{(100)}^{(1)}$ corresponding to the large period $P_{(100)}^{(1)}$ in (b) is given by $Q_{(100)}^{(1)} = (4\pi/a) - Q_{\text{belly}}$ where Q_{belly} is the belly radius along the ΓX direction in the Brillouin zone and a the lattice constant.

Unfortunately, there are no reported experimental results for oscillatory magnetic coupling across $\text{Cu}_{1-x}\text{Ni}_x$ spacers for the (100) direction. The importance of this direction lies in the fact that two different oscillation periods are present both in theory and experiment for pure Cu [14] and they both originate from extremal vectors at the belly of the Fermi surface. In particular, as can be seen in figure 1(a), the two extremal vectors are the vertical dimensions of the ‘dog’s bone’. There is again significant agreement between theory and experiment for pure Cu, in spite of the extra complication of extracting the sizes of the two different oscillation periods from the experimental data. In figure 3(b) we have plotted the

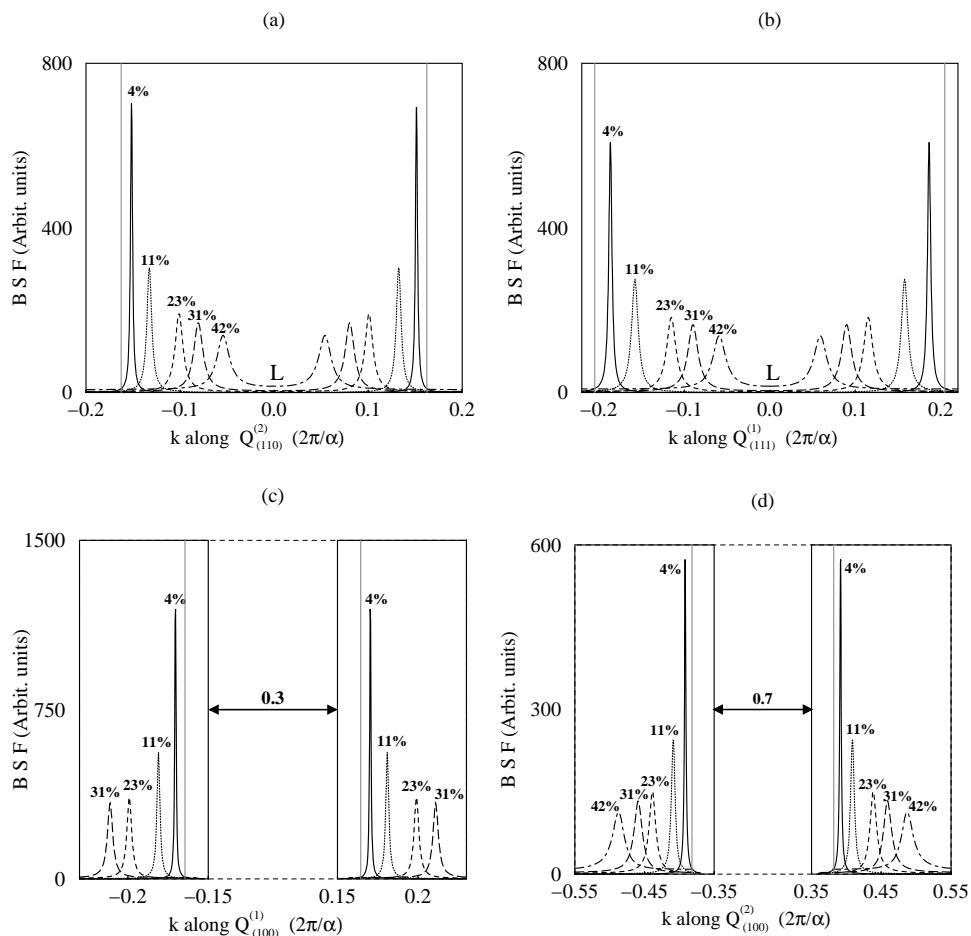


Figure 4. The Bloch spectral functions for different concentrations along the extremal vectors $Q_{110}^{(2)}$ (a), $Q_{111}^{(1)}$ (b), $Q_{100}^{(1)}$ (c), $Q_{100}^{(2)}$ (d) shown in figure 1. The separation of the two peaks is the length of the extremal vector, while the halfwidth of the Lorentzian-like peaks is the inverse mean free path. The vertical grey lines indicate the positions of delta function peaks for pure Cu. Notice that in all cases the Fermi surface is well defined. Inverting the Q -vectors and the halfwidths we get the corresponding oscillation periods and mean free paths with the second being much larger than the first.

calculated oscillation periods for the (100) direction as functions of the Ni concentration. The amplitude of the large-period oscillation in the case of pure-Cu spacer has been found to be much smaller than that of the small-period ones [14, 21–23, 30] and it has been impossible in some experiments [30] to observe the large-period oscillation at all. The extremal vector corresponding to the small oscillation (which is dominant) connects two points close to the areas of two different necks, as can be seen in figure 1(a). Thus, it is worth noticing that all of the clearly observed periods originate from extremal vectors with their end-points close to the area of the necks of the Fermi surface. However, the size of the large period corresponds to the size of the (100) belly radius of the Cu Fermi surface and it would be interesting to observe the analogous period for $\text{Cu}_{1-x}\text{Ni}_x$ alloy spacers.

In the light of the above discussion it is evident that at least for $\text{Cu}_{1-x}\text{Ni}_x$ binary alloy

spacers the oscillation periods correspond to extremal vectors of the alloy Fermi surface like in the pure-Cu case. There is excellent agreement between theory and experiment for the evolution of the periods with concentration and the degree of agreement does not seem to be influenced by alloying.

Finally, we will discuss the question of oscillation damping in the case of coupling across alloy spacers. We have already mentioned that for those binary alloy spacers for which the Fermi surface of the bulk alloy is well defined we do not expect to see significant damping of the oscillations in the range of the first few oscillation periods. Equation (2) implies that the damping of the oscillations is an entirely local effect, i.e. it depends exclusively on the mean free paths of the quasiparticle states at the end-points of the extremal vector of the Fermi surface which corresponds to the particular oscillation. Usually, the extremal vectors of interest connect two equivalent points on the Fermi surface, so the oscillation damping depends on the mean free path at a particular point of the Fermi surface projected in the direction of growth. Thus, the sufficient condition for not observing oscillation damping in the range of the first few periods is for the Fermi surface of the binary alloy to be well defined locally. For example, if the extremal vector related to an oscillation spans a neck or pocket of the Fermi surface, it is sufficient that the inverse mean free paths, a measure of which are the halfwidths of the Lorentzian-like spectral function peaks, be small in comparison to the dimensions of the neck or pocket, i.e. the size of the extremal vector. Other parts of the Fermi surface might be smeared by disorder. A related discussion on the consistency of the calculated mean free paths from the BSF with the lack of any observation of oscillation damping in the oscillatory magnetic coupling as well as the residual resistivity of alloys is reported in reference [19]. In the present work we think it useful to show some calculated BSF for different concentrations along the directions of the extremal vectors involved in the oscillatory coupling across $\text{Cu}_{1-x}\text{Ni}_x$ binary alloy spacers. In figure 4 the BSF along the direction of the extremal vectors is plotted, for $Q_{(110)}^{(2)}$ (neck diameter), $Q_{(111)}^{(1)}$, $Q_{(100)}^{(1)}$ (ΓX belly radius) and $Q_{(100)}^{(2)}$. The position of the peaks for each concentration indicates the end-points of the extremal vector, with their distance being the size of the extremal vector.

We see that in more or less all of the cases the halfwidth of the peaks is indeed small compared with the size of the vector. Even the small-size neck is well defined (figure 4(a)) for all of the concentrations examined. In all cases, this result is in agreement with either the experiment [25–27], where antiferromagnetic peaks are observed for large distances for (111) and (110) growth directions, or theoretical total-energy calculations [31] for the (100) direction where also no damping was observed or inferred.

4. Conclusion

In conclusion, we have presented theoretical calculations for $\text{Cu}_{1-x}\text{Ni}_x$ alloy Fermi surfaces and shown them to be in very good agreement with oscillatory magnetic coupling experiments across $\text{Cu}_{1-x}\text{Ni}_x$ alloy spacers. In particular we find an excellent correspondence between the observed periods and the calculated extremal vectors on the alloy Fermi surface. On the other hand, however, the oscillation amplitude dependence on the spacer thickness does not seem to be affected by alloying, in so far as the alloy Fermi surface is well defined. Thus the oscillation damping expected in the alloy spacer case is not significant within the first few observed oscillation periods as a result of the very large mean free paths of the quasiparticle states at the Fermi surface. In other words, as soon as the binary alloy Fermi surface is well defined, the characteristic length of the damping is much larger than the oscillation period. The agreement between the calculated and measured

characteristics of the alloy Fermi surface suggests that oscillatory magnetic coupling across alloy spacers may become a very important and challenging technique, which along with the ACAR method could be applied to the study of the Fermi surfaces of random binary alloys.

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