

# Magnetically mediated superconductivity in heavy fermion compounds

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**In a conventional superconductor, the binding of electrons into the paired states that collectively carry the supercurrent is mediated by phonons—vibrations of the crystal lattice. Here we argue that, in the case of the heavy fermion superconductors  $\text{CePd}_2\text{Si}_2$  and  $\text{CeIn}_3$ , the charge carriers are bound together in pairs by magnetic spin–spin interactions. The existence of magnetically mediated superconductivity in these compounds could help shed light on the question of whether magnetic interactions are relevant for describing the superconducting and normal-state properties of other strongly correlated electron systems, perhaps including the high-temperature copper oxide superconductors.**

The ancient observation of action at a distance between magnetic materials such as lodestone inspired William Gilbert to postulate an analogous force between the Earth and the Sun<sup>1</sup>. Later work showed that a parallel force holds atoms together. This charge–charge interaction is also ultimately responsible, in a subtle way, for the binding of Cooper pairs in superconductors. In traditional superconductors<sup>2</sup>, this binding is most simply described in terms of the emission and absorption of waves of lattice density. Here we consider instead whether this binding could in some materials be most simply described in terms of the emission and absorption of waves of the electron spin density: that is, whether a type of bound state that was only identified this century can arise because of an effective spin–spin interaction (see, for example, refs 3–7) that is reminiscent of one of the oldest known forces.

The low-temperature properties of interacting electrons in conductors are normally described in terms of excited states known as ‘quasiparticles’<sup>3,9</sup>. These entities are liable to be very different from bare electrons. Although they may obey the same ‘fermion’ quantum statistics, they can behave as though they are very ‘heavy’. We are interested in materials that support such quasiparticles, namely the ‘heavy fermion’ compounds. Exotic interactions can take place between quasiparticle charge carriers via their host medium, leading to new and subtle states of matter. One may imagine that, at each instant in its motion, a quasiparticle emits a wave that perturbs its environment in the manner of a stone entering a pond. Another charge carrier elsewhere can receive the signals, and thus charge carriers are able to communicate with each other. It is possible to tune the nature of these communications by altering the density of the crystal. In this way it is possible to alter dramatically the transport behaviour and other measured properties of a material. And in sufficiently calm conditions at low temperatures, bound states may form leading, in particular, to superconductivity.

We consider whether it is possible to tune the nature of charge carrier interactions such that magnetic coupling demonstrably dominates the more conventional non-magnetic channels of communication. In particular, we examine whether in such a scenario it would be possible for a magnetically mediated form of superconductivity to exist at sufficiently low temperatures. We present new evidence to suggest that this kind of superconductivity may indeed exist in some heavy fermion compounds, but that it is normally only viable extremely close to the critical lattice density  $n_c$

at which long-range magnetic order is suppressed, and in specimens of extremely high purity. We discuss a magnetic-interactions model that provides a natural description of the temperature–density phase diagrams we have observed in two particularly simple heavy fermion compounds, namely cubic  $\text{CeIn}_3$  and tetragonal  $\text{CePd}_2\text{Si}_2$ . In each of these two systems, we find that both the variation of the magnetic and superconducting transition temperatures with lattice density, and the anomalous forms of the normal state resistivities, are qualitatively consistent with the magnetic-interactions model, which uses independently estimated parameters. This agreement, together with other features described here, provides compelling evidence for the probable existence of magnetically mediated superconductivity. Moreover, the magnetic interactions model, which may account for superconductivity in the materials studied, suggests how the properties of a material might be modified in order to increase the superconducting transition temperature. Interestingly, the changes required would lead us to materials with some of the features of the copper oxide superconductors.

## The edge of magnetic order

We consider how to tune the charge carrier interactions so that the magnetic channel may become dominant. During the course of its motion, a quasiparticle nucleates various waves in the medium which affect other quasiparticles. The amplitudes of such waves depend on the strength of the coupling of a quasiparticle with the medium, and on the ease with which such waves can be excited in the medium. Magnetic waves of interest may be readily excited when a magnetic medium is near a critical lattice density,  $n_c$ , that renders it close to entering a state of continuous long-range magnetic order. Such waves in the non-magnetically ordered state are usually damped and may be described in terms of a relaxation frequency spectrum,  $\Gamma_{\mathbf{q}}$ . And this quantity, which describes the characteristic rate of decay of a spontaneous fluctuation of the magnetization of wavevector  $\mathbf{q}$ , characterizes the dynamics of quasiparticle interactions (for a review, see ref. 10; see also refs 11–14).

In the simplest model, the retarded interaction produced by one quasiparticle on another depends on the product of three factors<sup>4</sup>, namely the relative orientation and magnitudes of the magnetic moments involved,  $-\boldsymbol{\mu}_1 \cdot \boldsymbol{\mu}_2$ , the square of a coupling parameter, and the space and time-dependent magnetic susceptibility, which itself depends on  $\Gamma_{\mathbf{q}}$ . When it is dominant, this interaction can lead to two striking consequences. First, near  $n_c$ , magnetic waves tend to propagate over a long range. If the coupling parameter for such waves remains finite, the magnetic interactions are therefore also

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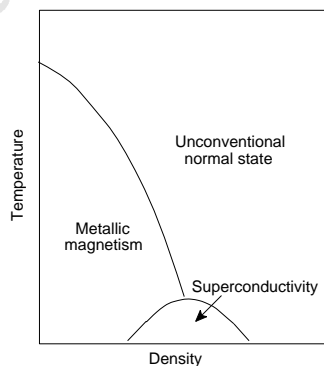
long-range. This can lead, as we shall see, to properties that are very different from those normally associated with metals, or with the so-called Fermi liquid model of the normal metallic state<sup>8,9</sup>. Second, for some relative orientation of two quasiparticle moments, the interaction can be attractive in certain regimes of space and time. Hence bound states, and therefore superconductivity, may arise at low temperatures. In contrast to the classical superconductors, the bound states produced by magnetic interactions have orbital and perhaps spin angular momentum and are hence anisotropic in space. We also note that magnetic interactions tend to interfere with pairing via phonons and therefore are usually expected to suppress conventional forms of superconductivity in nearly magnetic metals<sup>15,16</sup>. Further, we note that the coupling parameter may be non-local and that the response of the medium may be nonlinear. Hence in general magnetic interactions may not be described by the space-time susceptibility alone. The magnetic interactions model leads to a qualitative phase diagram of the form shown in Fig. 1.

**The choice of materials**

Evidence for the above phase diagram may be sought in compounds in which (1) the critical density  $n_c$  may be reached with readily accessible pressures (in our case <30 kbar); (2) the states are metallic on both sides of  $n_c$  with a single magnetic transition near  $n_c$ , or otherwise other interaction channels are liable to be important; (3) the magnetic transition is continuous at  $n_c$ , or otherwise the magnetic interactions would not be long-range nor necessarily dominant; and (4) pair-breaking effects, in particular impurity scattering, can be made sufficiently small.

Simple candidates include  $d$  metal ferromagnets<sup>17–24</sup>, but these suffer from problems of purity, remoteness from  $n_c$  or, in some cases, pair-breaking due to lack of inversion symmetry. Similar complications arise in simple  $d$  metal antiferromagnets in which, moreover, the magnetic transitions are often not continuous. More promising candidates can be found among the  $f$  metals. We consider in particular the cerium-based heavy fermion systems.

To narrow the list down further, we examine the prediction of the magnetic interactions model for the superconducting transition temperature  $T_c$ . The solutions of the Eliashberg equations suggest that  $T_c \sim \Gamma \theta \phi \exp(- (1 + \lambda)/g\lambda)$ ; (see ref. 6 for example). Here  $\Gamma$  is the bandwidth of the spectrum  $\Gamma_q$ ,  $\lambda$  represents the enhancement of the quasiparticle mass due to magnetic interactions<sup>10–14</sup> which



**Figure 1** Possible temperature–density phase diagram of a pure metal in which magnetic order is quenched gradually with increasing lattice density. Near the critical density  $n_c$ , where the magnetic transition temperature vanishes, magnetic interactions become strong and long-range. The normal state is expected to be anomalous here and, at sufficiently low temperatures, it is expected to give way to a kind of superconductivity in which Cooper pairs are bound together by a glue of magnetic origin. Superconductivity may exist only over a very narrow range of densities near  $n_c$ —which is where magnetic interactions overwhelm other channels. Moreover, superconductivity may exist only in samples in which the carrier mean free path exceeds the superconducting coherence length. In most cases this requires samples of very high purity.

depends on a  $\mathbf{q}$ -space average of  $1/\Gamma_q$ , and  $g$  measures the relative strength of the interaction in the given pair state. Nearly ferromagnetic metals are expected to produce pairs in spin-triplet states with an odd orbital angular momentum quantum number  $l$ , whereas nearly antiferromagnetic metals should produce spin-singlet states with  $l$  even (and normally greater than zero)<sup>3–7</sup>. The factor  $\theta$  depends on the form of  $\Gamma_q$  and, in part, represents damping due to incoherent inelastic scattering;  $\phi$  represents damping due to scattering from impurities (either magnetic or non-magnetic for anisotropic pair states): it falls when there is an increase in the ratio of the superconducting coherence length  $\xi$  to the carrier mean free path  $l_{mfp}$ .

As it stands, this model for  $T_c$  predicts that superconductivity should be widespread in pure cerium-based heavy fermion compounds. But, despite an extensive search for two decades, superconductivity has been found at ambient pressure in these systems only in CeCu<sub>2</sub>Si<sub>2</sub> (ref. 25). Here,  $T_c$  is insensitive to pressure and the pairing mechanism may be different from that described above<sup>26,27</sup>. Indeed, the collapse of  $T_c$  upon reaching  $n_c$  in the closely related system CeCu<sub>2</sub>Ge<sub>2</sub> under pressure<sup>28</sup> might well be described in terms of the pair breaking effects of magnetic interactions, as suggested in the early proposals regarding ambient pressure Pd (refs 15, 16).

The scarcity of superconductivity in the cerium heavy fermion metals may be due to impurities and competing channels. Sufficiently close to  $n_c$  however, magnetic interactions become long-range and can dwarf competing channels, at least for pair states of sufficiently high  $l$ . Moreover, because the quasiparticle mass  $m^*$  grows as  $n_c$  is approached<sup>10–14</sup>,  $\xi$  tends to a minimum and hence  $\phi$  tends to a maximum—at least if  $l_{mfp}$  does not fall too rapidly as  $n \rightarrow n_c$ . On the other hand,  $\theta$  drops near  $n_c$ , and may indeed collapse in the nearly ferromagnetic metals<sup>5</sup>. This effect is less extreme in nearly antiferromagnetic metals and  $T_c$  is a maximum at  $n_c$  (ref. 29). We therefore confine our search to antiferromagnetic heavy fermion systems.

We estimate that in contrast to the  $d$  metals, it is possible to produce sufficiently pure cerium-based systems because  $\xi$  tends to be short. After an extensive survey, it was decided that CePd<sub>2</sub>Si<sub>2</sub> and CeIn<sub>3</sub> were among the materials most likely to satisfy all the above criteria. CePd<sub>2</sub>Si<sub>2</sub> crystallizes in a body-centred tetragonal structure like CeCu<sub>2</sub>Si<sub>2</sub> and is, at ambient pressures, an antiferromagnet with  $\mathbf{Q}$  along [110] below a Néel temperature  $T_N$  of  $\sim 10$  K (refs 30–33). CeIn<sub>3</sub> forms a simple cubic structure and at ambient pressure orders in a classical antiferromagnetic state with  $\mathbf{Q}$  along [111] below  $T_N \sim 10.1$  K (refs 34–36). In both systems, the critical density  $n_c$  (where  $T_N$  vanishes) can be reached with an applied pressure in the range 25 to 30 kbar (refs 33, 36).

**Measurements of CePd<sub>2</sub>Si<sub>2</sub> and CeIn<sub>3</sub>**

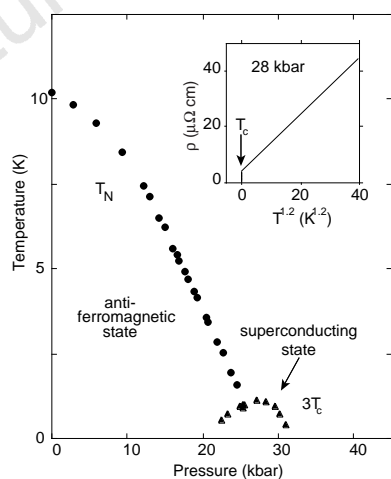
High-purity samples of CePd<sub>2</sub>Si<sub>2</sub> were synthesized as follows. Reactants (Ce 99.999%, Pd 99.9985%, Si 99.999%) were heated by the non-invasive technique of radiofrequency induction on a water-cooled copper boat in an ultrahigh-vacuum system which could be pressurized to up to 5 atmospheres with ultrahigh-purity argon after *in situ* purification with a hot cerium getterer (the starting purity was better than 1 p.p.m.). Our  $a$ -axis resistivity measurements were made on  $a$ – $b$  plane single-crystal platelets from the resulting annealed ingot, the best of which had a residual resistivity of a few  $\mu\Omega$  cm, which is a factor of five below values previously reported. Electron microprobe analysis revealed neither any evidence of inhomogeneities (to a resolution of 0.1%), nor the presence of a second phase. A similar procedure was employed in the synthesis of CeIn<sub>3</sub>.

Single-crystal resistivity measurements under pressure were made by a low-power a.c. four-terminal method within conventional BeCu/maraging steel piston-cylinder cells, filled with an equal mixture of *iso*-pentane and *n*-pentane<sup>17</sup>. The pressure was determined to  $\pm 0.1$  kbar from the superconducting transition of Sn, and the temperature via Ge and RhFe sensors placed on a Cu enclosure

in good thermal contact with both the pressure cell and the sample leads. To try to ensure adequate pressure and temperature homogeneity, a slow cooling rate from room temperature, typically  $0.2 \text{ K min}^{-1}$ , was employed. Measurements were carried out from room temperature to the millikelvin temperature range within a pumped  $^4\text{He}$  cryostat, an adiabatic demagnetization refrigerator and a top-loading dilution refrigerator.

Our key experimental results are summarized in Figs 2, 3. In the materials studied, the antiferromagnetic ordering temperature  $T_N$ , at which there is a discontinuity in the gradient of the resistivity  $\rho$  (not shown), was found to decrease slowly and monotonically with increasing pressure,  $p$ . Over a wide region of the  $\text{CePd}_2\text{Si}_2$  phase diagram,  $T_N$  is close to being linear in  $p$ , and extrapolation from this regime to absolute zero allows us to define an effective critical pressure  $p_c$  of 28 kbar. In  $\text{CeIn}_3$ , the variation of  $T_N$  with  $p$  is more rapid, and we estimate  $p_c$  to be  $\sim 26$  kbar. The behaviour of  $T_N$  as it falls below 1 K has not been resolved in these studies.

In the case of  $\text{CePd}_2\text{Si}_2$ , the resistivity  $\rho$  does not exhibit the standard  $T^2$  form expected of a Fermi liquid. Careful analysis shows that near  $p_c$  it in fact varies as  $T^{1.2 \pm 0.1}$  over nearly two decades in temperature down to the millikelvin range (Fig. 2, inset). Below 500 mK and in a narrow region near  $p_c$ , we observe an abrupt drop in  $\rho$  to below the detection limit, consistent with the occurrence of a superconducting transition, as discovered during our initial observations in September 1994<sup>37,38</sup>. At a given pressure, this transition may be characterized by a temperature  $T_c$ , at which  $\rho$  falls to 50% of its normal state value. The width of this transition grows markedly as the pressure is varied away from  $p_c$ . We stress that experimentally,  $\rho$  is found to actually vanish only close to  $p_c$ . By energizing a Nb–Ti coil placed in our pressure cell, it was established that the upper critical field  $B_{c2}$  varies as  $dB_{c2}(p_c)/dT \sim -6 \text{ T/K}$  near  $T_c$ . This is a high rate of change for such a small value of  $T_c$ —much higher than the expected figure for a conventional superconductor. However, it is the same order of magnitude as the value found in the heavy fermion superconductor  $\text{CeCu}_2\text{Si}_2$  (ref. 27). We note that in a traditional analysis, the slope of  $B_{c2}(T)$  at  $T_c$  implies a superconducting coherence length of 150 Å, a value which is below the value of  $l_{\text{mfp}}$  that we estimate for our best samples. No superconductivity has been observed in specimens with residual resistivities above several  $\mu\Omega \text{ cm}$ , namely those with an estimated  $l_{\text{mfp}}$  that is



**Figure 2** Temperature–pressure phase diagram of high-purity single-crystal  $\text{CePd}_2\text{Si}_2$ . Superconductivity appears below  $T_c$  in a narrow window where the Néel temperature  $T_N$  tends to absolute zero. Inset: the normal state  $\rho$ -axis resistivity above the superconducting transition varies as  $T^{1.2 \pm 0.1}$  over nearly two decades in temperature<sup>27,30</sup>. The upper critical field  $B_{c2}$  at the maximum value of  $T_c$  varies near  $T_c$  at a rate of approximately  $-6 \text{ T/K}$ . For clarity, the values of  $T_c$  have been scaled by a factor of three, and the origin of the inset has been set at 5 K below absolute zero.

substantially below  $\xi$  (for a similar example, see ref. 39).

In the case of cubic  $\text{CeIn}_3$ , we find that very close to  $p_c$  the normal state resistivity assumes a non-Fermi liquid form, but this time<sup>40,41</sup> varies as  $T^{1.6 \pm 0.2}$ . Thus, near their respective critical pressures, the resistivity exponent in the cubic material is significantly higher than it is in tetragonal  $\text{CePd}_2\text{Si}_2$ . In a very narrow region near  $p_c$ , we again see a sharp drop in  $\rho$  to below the detection limit, but at somewhat lower temperatures than the transitions observed in  $\text{CePd}_2\text{Si}_2$ . This is consistent with the occurrence of superconductivity in yet another cerium compound on the edge of long-range magnetic order<sup>40,41</sup>.

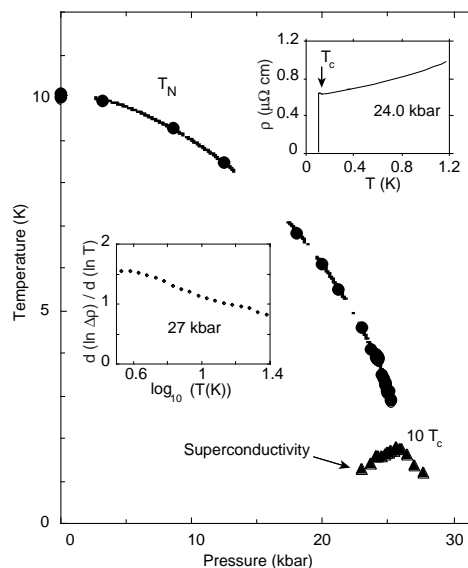
We stress that in each material studied, both the form of the temperature dependence of the normal state resistivity, and the nature and existence of the superconducting transition are sensitive to sample quality. In particular, the superconducting transitions appear only in samples with residual resistivities in the low  $\mu\Omega \text{ cm}$  range, as expected in the case of anisotropic pair states with coherence lengths of the order of a few hundred ångströms.

### Magnetic interactions

The observed temperature–pressure phase diagrams for both  $\text{CePd}_2\text{Si}_2$  and  $\text{CeIn}_3$  are at least qualitatively consistent with what is expected in terms of the magnetic interactions model (Fig. 1). We now consider a more quantitative comparison. In the following it is assumed that the magnetic transition is continuous and that  $n$  is close to  $n_c$ . The incoherent scattering of quasiparticles via magnetic interactions is then expected to lead to a resistivity of the form

$$\rho = \rho_0 + AT^x \quad (1)$$

where  $\rho_0$  and  $A$  are constants and the exponent  $x$  is smaller than two, that is, smaller than it is in a conventional Fermi liquid at low  $T$



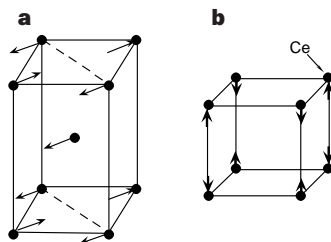
**Figure 3** Temperature–pressure phase diagram of high-purity single-crystal  $\text{CeIn}_3$ . A sharp drop in the resistivity consistent with the onset of superconductivity below  $T_c$  is observed in a narrow window near  $p_c$ , the pressure at which the Néel temperature  $T_N$  tends to absolute zero. Upper inset: this transition is complete even below  $p_c$  itself. Lower inset: just above  $p_c$ , where there is no Néel transition, a plot of the temperature dependence of  $d(\ln \Delta\rho)/d(\ln T)$  is best able to demonstrate that the normal state resistivity varies as  $T^{1.6 \pm 0.2}$  below several degrees K (ref. 29) ( $\Delta\rho$  is the difference between the normal state resistivity and its residual value—which is calculated by extrapolating the normal-state resistivity to absolute zero). For clarity, the values of  $T_c$  have been scaled by a factor of ten. The resistivity exponents of  $\text{CeIn}_3$  and  $\text{CePd}_2\text{Si}_2$  may be understood by taking into account the underlying symmetries of the antiferromagnetic states and using the magnetic interactions model. Superconductivity near  $n_c$  in pure samples is expected to be a natural consequence of the same model.

(refs 10, 11, 14, 42). For a three-dimensional ferromagnet,  $x = 5/3$ , in agreement with several nearly ferromagnetic  $d$  metals<sup>17,21</sup>. For an antiferromagnet,  $x = 3/2$  in three dimensions and  $x = 1$  in two dimensions<sup>11,14</sup>, provided that in both cases  $\rho_0$  and  $T$  are not too low<sup>43</sup>. The former is indeed consistent with our findings in cubic CeIn<sub>3</sub>, whereas the latter is closer to tetragonal CePd<sub>2</sub>Si<sub>2</sub>. The quasi two-dimensional character of the spin-relaxation spectrum of CePd<sub>2</sub>Si<sub>2</sub> may be anticipated from the observed [110] direction of the ordering wavevector  $\mathbf{Q}$  observed at ambient pressure. As shown in Fig. 4, the spin configuration for this case implies that the nearest-neighbour spin coupling along the  $c$  direction is frustrated. This leads to a weaker variation of  $\Gamma_{\mathbf{Q}+\mathbf{q}}$  when  $\mathbf{q}$  is along the  $c$  axis than when it lies in the basal plane, and hence to a quasi two-dimensional behaviour for the magnetic interactions. We note that along the  $c$  axis, if a slow  $q^4$  variation in  $\Gamma_{\mathbf{Q}+\mathbf{q}}$  survives in an even-power expansion of  $\Gamma_{\mathbf{Q}+\mathbf{q}}$  in  $q$ , then the effective dimensionality is  $5/2$  and  $x = 5/4$ . In this way, the exponent  $3/2 > x > 1$  observed in CePd<sub>2</sub>Si<sub>2</sub> can be interpreted naturally. But because of the reduced dimensionality, an elementary treatment may in this case be insufficient.

The magnetic interactions model also predicts that the magnetic transition temperature collapses as  $|p - p_c|^\gamma$  near  $p_c$  (refs 10–14). For a three-dimensional ferromagnet, the exponent  $\gamma = 3/4$ , in agreement as before with observations in the  $d$  metals<sup>17,21</sup>. For an antiferromagnet,  $\gamma = 2/3$  in three dimensions, and  $\gamma = 1$  in two dimensions. The quasi-linear variation of  $T_N(p)$  in CePd<sub>2</sub>Si<sub>2</sub> and the more rapid  $T_N(p)$  in CeIn<sub>3</sub> near  $p_c$  are not inconsistent with these predictions.

At sufficiently low temperatures, the incoherent scattering that yields  $\rho(T)$  is expected to go over to coherent scattering if the quasiparticles attract. This can lead to bound states and superconductivity. We now examine the predicted order of magnitude of  $T_c$ . Near  $n_c$ , magnetic interactions are expected to dominate other channels and for our samples,  $\phi \approx 1$ . In these circumstances, the numerical solutions of the Eliashberg equations for the  $T_c$  of antiferromagnets given in ref. 29 may be relevant. At  $n_c$ , the expected  $T_c$  depends strongly on  $\Gamma$ , which may be estimated from bulk properties and neutron-scattering data<sup>30–32,34–36</sup>. In both CeIn<sub>3</sub> and CePd<sub>2</sub>Si<sub>2</sub>, we find  $\Gamma$  to be 30–50 K near  $p_c$ . Ref. 29 then suggests maximum values of  $T_c$  of a few tenths of a degree K at  $n_c$ , in order of magnitude agreement with our observations. With the same parameters, we can also understand the order of magnitude of  $A$  in equation (1) (ref. 41).

The estimates of  $T_c$  apply only very close to  $n_c$ . In impure samples, the extremely rapid fall of  $T_c(p)$  may be due to a drop in  $\phi$ , whereas in pure samples, an anticipated sharp fall in  $g\lambda$  and the contribution of other interaction channels may take over. These effects may naturally account for the difficulty of finding magnetically mediated superconductors.



**Figure 4** The positions and spin alignments of the Ce atoms in the unit cells of the two materials studied. **a**, The body-centred tetragonal unit cell of CePd<sub>2</sub>Si<sub>2</sub> has the antiferromagnetic ordering wavevector  $\mathbf{Q}$  along [110]. The Pd and Si atoms are not shown. Note that, by symmetry, the magnetic couplings along the  $c$  axis between nearest neighbours would actually vanish in this spin configuration. **b**, The simple cubic unit cell of CeIn<sub>3</sub> has  $\mathbf{Q}$  along [111]. The In atoms (not shown) are located at the centre of the faces of the cubic unit cell.

### Link to other highly correlated fermion systems?

Further evidence for the existence of magnetically mediated superconductivity in cerium heavy fermion compounds arose when similar findings were reported in CeRh<sub>2</sub>Si<sub>2</sub> (ref. 44) and CeCu<sub>2</sub> (ref. 45). It is possible that these materials may also be understood in terms of the magnetic interactions model discussed here. But a detailed analysis is not yet possible. The metallurgical properties of CeRh<sub>2</sub>Si<sub>2</sub> and CeCu<sub>2</sub> are complex and the transitions near  $n_c$  are more abrupt than those in CePd<sub>2</sub>Si<sub>2</sub> or CeIn<sub>3</sub>. Moreover, in CeRh<sub>2</sub>Si<sub>2</sub> and possibly CeCu<sub>2</sub> as well, there exist two or more magnetic transitions that considerably complicate theoretical modelling. More recently, superconductivity has also been observed in CeNi<sub>2</sub>Ge<sub>2</sub><sup>46,47</sup>. This system has the same lattice and electronic structure as CePd<sub>2</sub>Si<sub>2</sub> and behaves at ambient pressure much as CePd<sub>2</sub>Si<sub>2</sub> does at  $p_c$ . Thus, there is increasing evidence, since the early observations in CePd<sub>2</sub>Si<sub>2</sub> and CeRh<sub>2</sub>Si<sub>2</sub>, that superconductivity may occur quite generally in the immediate vicinity of  $n_c$ , in samples of sufficiently high quality, at least when the magnetic transition is continuous.

These considerations may also be important in the uranium-based heavy fermion superconductors<sup>48–53</sup>. But in some of these systems, magnetic interactions may compete with others that are associated with incipient quadrupolar or higher multipolar ordering<sup>52,53</sup>. Similarly, superconductivity observed on the border of metal–insulator Mott transitions may also originate from magnetic binding<sup>9,54–56</sup>. But the magnetic transitions may not be continuous and moreover, subtle interactions associated with incipient localization could be important in these systems<sup>9,56</sup>. Magnetic couplings are also thought to arise in liquid <sup>3</sup>He and play a key role in the formation of the triplet  $p$ -wave superfluid state below a few millikelvin (refs 3, 4, 57). Even in this relatively simple fermionic system, however, the quasiparticle interactions may be complex and, in particular, may reflect the tendency of the quantum liquid to order in magnetic and spatially localized states.

It is interesting to consider how the low transition temperatures of the cerium based heavy fermion superconductors might be increased within the framework of the present magnetic pairing model. The formula presented earlier for  $T_c$  suggests that it is necessary to increase the bandwidth  $\Gamma$  of the magnetic relaxation spectrum without simultaneously forcing a drastic reduction in the parameter  $\lambda$  which appears in the exponent. The bandwidth may be increased by going over from narrow  $f$  band materials to broader  $d$  band systems—and  $\lambda$ , which depends on a momentum–space average of  $1/\Gamma_{\mathbf{q}}$  (refs 10–14), may be prevented from dropping excessively by reducing the dispersion along one axis, by reducing the effective dimensionality. This also has the advantage of generally increasing  $g$ , particularly in the anisotropic singlet case. In addition, we note that for spin 1/2 particles, the magnitude of  $\boldsymbol{\mu}_1 \cdot \boldsymbol{\mu}_2$ , which appears in the formula for  $T_c$ , has a quantum average that is three times larger in the singlet state than it is in the triplet state. Both this fact, and the effects of incoherent scattering, tend to favour antiferromagnetic rather than ferromagnetic interactions.

The sum of these considerations would lead us to look for high superconducting transition temperatures in  $d$  metal compounds of reduced dimensionality on the border of antiferromagnetic order. It is interesting that these properties are in fact similar to those of the copper oxide superconductors. This may prove to be a coincidence, as other factors beyond those given here may be of comparable or even greater importance. Nevertheless, the search for a possible interpretation of the behaviour of the copper oxides in terms of some sort of magnetic-interactions model seems reasonable on the basis of the findings summarized here. Several scenarios with similar starting outlooks to the one we describe<sup>3–7,65–67</sup> have been put forward<sup>58–61,68</sup>, but even more exotic models may be required. As yet there is no general agreement on the best way of thinking about the copper oxides, but it is interesting that their spin liquid properties<sup>9,69</sup>, and—in common with our phase diagrams—the

interplay of antiferromagnetism and superconductivity<sup>62</sup>, are persistent and increasingly more frequent themes.

### 'Magnetic glue'

The superconductivity we have observed in the metals CePd<sub>2</sub>Si<sub>2</sub> and CeIn<sub>3</sub> is restricted to high-quality samples at the very edge of magnetic order. We believe that this is because it occurs only if there are attractive magnetic interactions that are strong enough to overcome competing interactions, and if it is possible for these magnetic interactions to create Cooper pairs that are small enough effectively not to see impurities. To support this statement, we argue that within the context of a single model based on magnetic interactions, it is possible to describe the key features of the phase diagrams and the anomalous normal state resistivities of the two materials in a consistent manner.

Therefore, we suggest that our evidence speaks strongly in favour of the existence of a superconducting state which arises because of magnetism rather than in spite of it. We imagine that the charge carriers in this superconducting state are held together in pairs by a 'magnetic glue'. Although magnetically mediated superconductivity is hard to prove because conditions must be just right in order to observe and infer its existence, we do not rule out the possibility that it is widespread. Thus our findings could shed some further light on current models of magnetic coupling in other strongly correlated electron systems—extending perhaps even to the high-temperature superconductors. If there is an alternative interpretation of our experimental results, it would probably involve even more unconventional thinking.

*Note added in proof:* Evidence in support of the 5/2 dimensional model discussed for CePd<sub>2</sub>Si<sub>2</sub> (ref. 46) has been found via recent inelastic neutron scattering<sup>63</sup> in CeCu<sub>6-x</sub>Au<sub>x</sub> (ref. 64). In contrast to CePd<sub>2</sub>Si<sub>2</sub> and CeIn<sub>3</sub>, this system is disordered and hence does not exhibit the form of the resistivity or the superconductivity discussed in this Article. □

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