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Influence of the spin susceptibility on the peak effect in La-doped $CeRu_2$

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Abstract

The influence of doping the Laves-phase superconductor $CeRu_2$ with La is studied by resistivity and magnetization measurements. As the La concentration increases the electronic mean free path and the spin susceptibility decrease. These changes modify the pinning properties of the vortex lattice, especially the peak effect. Although the magnetic field, at which the vortex lattice undergoes a crossover from weak to strong pinning, is increased, the pinning potential itself is not affected by the impurities. © 1998 Published by Elsevier Science B.V. All rights reserved.

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1. Introduction

The superconducting Laves phase compound $CeRu_2$ has attracted significant attention during the last few years. Starting point was the experimental observation [1] of a peak in the magnetization of this 6.1 K superconductor, with a large hysteresis for fields between about 0.7 H_{c2} and H_{c2}. Huxley et al. [2] suggested that this peak effect could be due to the formation of a new superconducting state that was predicted to exist many years ago by Fulde and Ferrell [3] and by Larkin and Ovchinnikov [4]. However, explanations in terms of the conventional peak effect [5] cannot be excluded.

After this first observation a large number of experiments were carried out and the theory was further developed. These works include: (i) the observation of the peak effect in heavy fermion superconductors like UPd_2Al_3 by Onuki et al. [6], who suggested that the effect is correlated to a large Pauli susceptibility; (ii) the claim of a first order transition from weak to strong pinning in superconductors with high spin susceptibility by Modler et al. [7]; (iii) flux flow transport measurements by Sato et al. [8] showing that the 'peak effect' exists up to the critical temperature; (iv) resistivity measurements showing the large critical current in the peak region [9]; and finally (v) vibrating reed experiments showing the importance of the vortex dynamics [10] and the strong softening of the crystal lattice in $CeRu_2$ [11,12].

The occurrence of the Fulde–Ferrel–Larkin–Ovchinnikov (FFLO) state is restricted by a number

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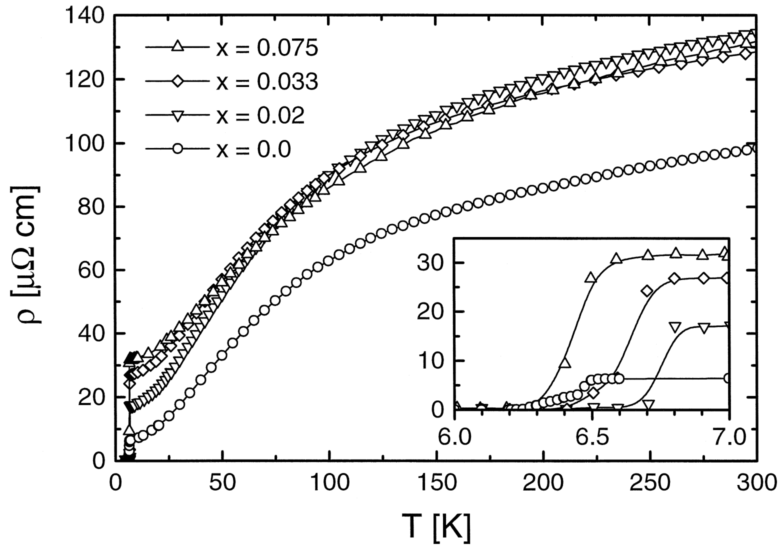

Fig. 1. Resistivity as a function of temperature of $Ce_{1-x}La_xRu_2$. The solid lines serve as a guide to the eye.

of criteria. First, the upper critical field must be Pauli-limited, i.e.,

$$\beta = \frac{\sqrt{2}\,H_{c2}(0)}{H_p} > 1.8,$$

and secondly, the superconductor should be in the clean limit, i.e., the mean free path has to be much larger than the coherence length. Burkhardt and Rainer [13] computed the phase diagram for the FFLO state in a two-dimensional system. They concluded that the FFLO state can only be stable below 0.56 T_c. Calculations by Takahashi et al. [14] and Tachiki et al. [15] removed this restriction by showing that a generalized FFLO state (GFFLO) may exist over the whole temperature region.

In this contribution we consider the effects of introducing nonmagnetic impurities into the system, thereby decreasing the mean free path l and thus violating the criterion for a clean superconductor. We have chosen La as the dopant material as opposed to a magnetic rare-earth dopant to avoid extra complications due to pair breaking effects.

2. Experimental

Four samples of $Ce_{1-x}La_xRu_2$ have been prepared by arc melting and subsequent annealing for 14 days at 1000°C. The samples were characterized by electron-probe microanalysis (EPMA). From this analysis it appeared that part of the La (about 30%) goes into a second phase, which then constitutes a few volume percent of the total sample and does not affect the bulk superconducting properties of the sample. Therefore, we will use the analyzed La concentration in the major phase to characterize the specimens: those are $x = 0.0$, 0.02, 0.033 and 0.075, respectively.

Electrical resistance was measured using a 4-probe ac-technique with an LR400 resistance bridge [2]; the magnetization was determined with a Quantum Design MPMS-5S SQUID magnetometer and ac-susceptibility was obtained using a standard mutual inductance bridge.

Fig. 1 depicts the electrical resistivity from 1.5 to 300 K measured on a small bar cut out of our samples. Clearly the overall behavior is the same for all samples except for the obvious increase in ρ_0 due to the La doping. The inset shows the low temperature part in detail. The superconducting transition temperature at first increases upon doping with La and then goes through a maximum at around $x =$

[2] Linear Research (San Diego, CA, USA).

Table 1
Electrical and magnetic properties of $Ce_{1-x}La_xRu_2$

Sample	T_c (K)	ρ_0 (μΩ cm)	l (Å)	χ_P ($\times 10^{-4}$)	$-(dH_{c2}/dT)T_c$ (T/K)
$x=0.0$	6.1	6	970	3.5	1.36
$x=0.02$	6.6	17	340	3.1	1.65
$x=0.033$	6.5	27	210	2.9	1.81
$x=0.075$	6.2	31	290	2.6	1.80
Sample	$\mu_0 H_{c2}(0)$ (T)	$\mu_0 H_p$ (T)	$\xi(0)$ (Å)	κ	β
$x=0.0$	5.8	7.8	75	28	1.1
$x=0.02$	7.6	8.8	66	34	1.2
$x=0.033$	8.2	9.0	63	38	1.3
$x=0.075$	7.8	8.9	58	38	1.3

0.02, in accordance with the observations reported by Shelton et al. [16] We will use the residual resistivity in the normal phase to deduce the mean free path. Note that this residual resistivity is only 6 μΩ cm for the pure $CeRu_2$. The values of ρ_0, T_c, and the mean free path l for all the samples can be found in Table 1.

After zero-field cooling, the magnetization of all samples was recorded during a field sweep of the SQUID in the so-called hysteresis mode using a scan length of 4 cm. This mode ensures a relatively quick measurement, which is necessary in view of the relaxation of the magnetization in the 'peak' [17]. The results were not affected by varying the scan length between 2 cm and 6 cm, showing that there is no detectable field inhomogeneity over those distances. Fig. 2 shows a typical result obtained on pure $CeRu_2$. We denote the field at which the onset of the peak becomes apparent as H^*. As also observed by many other workers, the peak closes again at or

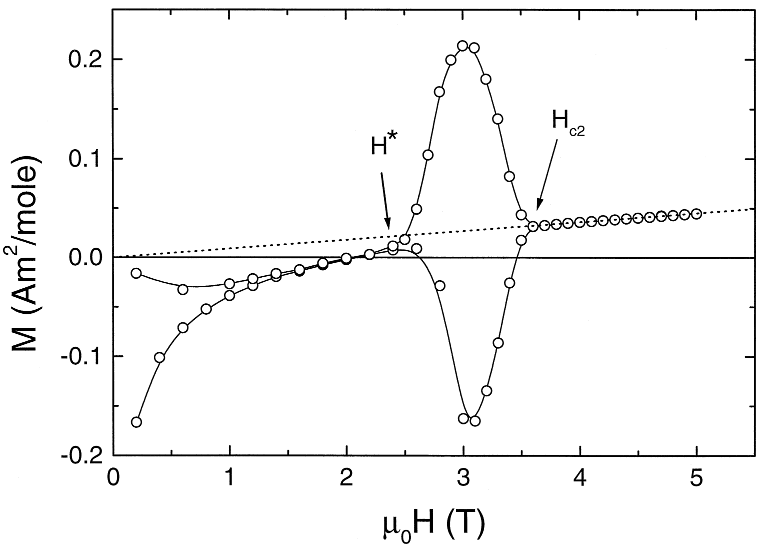

Fig. 2. Magnetization as a function of field for $CeRu_2$ at $T=4$ K after zero-field cooling. The dashed line represents the linear fit above H_{c2} from which the value for χ_P is extracted. H^*, the field at which the peak opens up, is indicated by the arrow.

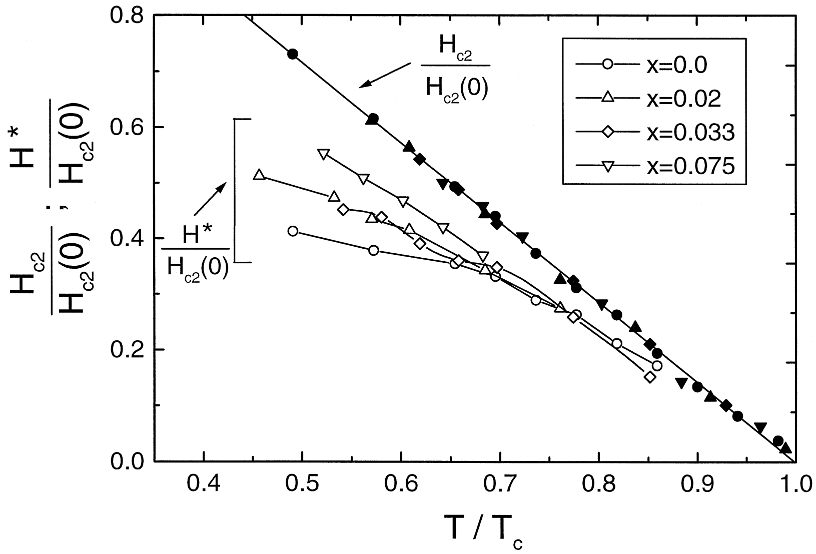

Fig. 3. Phase diagram of H^* (open symbols) and H_{c2} (closed symbols) versus temperature. Magnetic fields have been normalized with $H_{c2}(0)$.

slightly below H_{c2}. Above that field the magnetization is proportional to the field as indicated by the dashed line. From the slope of that line, we can determine the normal susceptibility χ_n of the samples. This is also the Pauli spin susceptibility χ_P of the normal electrons, assuming a negligible contribution [3] of the Landau diamagnetic susceptibility χ_L and a negligible Larmor susceptibility of the atomic cores. Similar curves were obtained for all samples up to a temperature of 5 K, while in ac-susceptibility experiments (not shown here) the peak-effect was observable up to 0.9 T_c. Note that our observation of the peak effect in La-doped samples is in disagreement with the results obtained by Roy and Chaddah [18].

3. Discussion

In Table 1, we collect the salient physical data characterizing our samples. The spin susceptibility χ_P decreases with increasing La concentration. At the same time, the field H^* shifts toward the upper critical field H_{c2}. This is shown in Fig. 3, where the field values have been normalized with respect to $H_{c2}(0)$ of the different samples in order to obtain a single $H_{c2}(T)$ line.

To establish the link between the spin susceptibility and the peak effect, one has to rule out the effect of the increased disorder (as indicated by the decreasing mean free path) on the pinning of the vortex lattice. The size of the peak gives a good indication for the pinning force density experienced by the vortex lattice. Now one is confronted with the problem of choosing the right temperature at which to compare the magnetization curves of the samples. Here we want to compensate for the effect of the spin susceptibility and study the influence of the increased disorder only. So comparing measurements at the same reduced temperature is misleading, because of the increase of the reduced field $h^*(T) = H^*(T)/H_{c2}(T)$ at which the peak opens up. We can compare the intrinsic pinning present in the samples by plotting the magnetization curves obtained at such temperatures that h^* has the same value for all samples. This is shown in Fig. 4(a) for $h^* = 0.8$ as a typical example. The size of the peak decreases with

[3] $\chi_L / \chi_P \propto (m/m^*)^2$ for free quasiparticles of spin 1/2 and effective mass m^*, which is enhanced in $CeRu_2$. See, e.g., N.W. Ashcroft and N.D. Mermin, *Solid State Physics*, Saunders College, Philadelphia, 1976, p. 666.

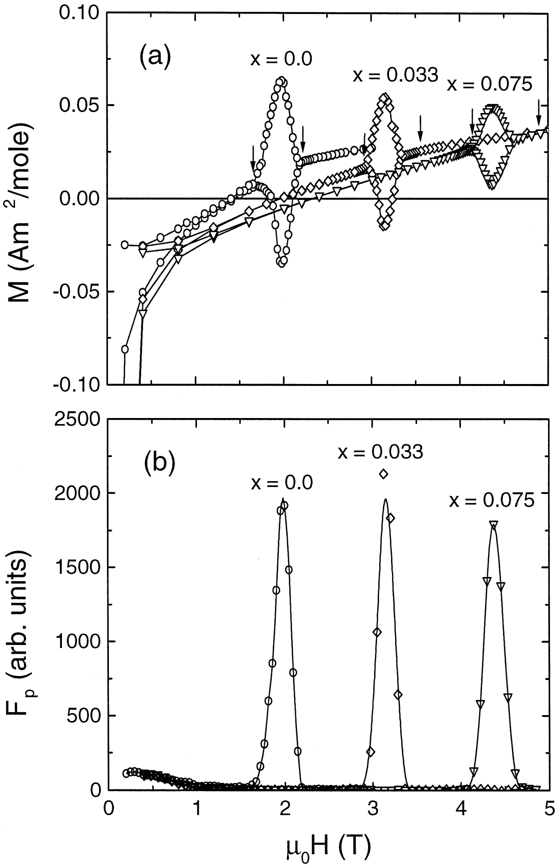

Fig. 4. (a) Magnetization curves after zero-field cooling at $T=4.5$ K ($x=0.0$), 4.5 K ($x=0.033$) and 3.5 K ($x=0.075$), at which $h^*=0.8$ as a typical example. The arrows indicate the fields H^* and H_{c2}. (b) Flux pinning densities obtained by multiplying the irreversible part of the magnetization curves (a) by the magnetic field. The solid lines serve as a guide to the eye.

increasing La concentration, but the position of the peak itself shifts towards higher magnetic fields. The pinning force density is estimated by multiplying the critical current (proportional to the difference ΔM in the magnetization between forward and reverse field cycles) with the magnetic field, see Fig. 4(b). We immediately see that it does not depend on La doping. By performing this comparison at other reduced fields h^* we always found the variations in F_p between the different samples to be less than 20%.

Thus, the intrinsic pinning potential present in the sample is not changed by the La-doping. The shift of H^* towards H_{c2} with increasing La concentration could be attributed to the lowering of the spin susceptibility. It has often been mentioned [2,6–8] that the enhanced spin susceptibility plays an important role in the properties of the vortex lattice in $CeRu_2$, through a large Zeeman energy term that counteracts the superconducting condensation energy. This term reduces the energy gain of pinning a single flux line. When considering individual vortices the pinning force density F_p is therefore expected to be lower for higher χ_P [19] [4], in disagreement with our results. Indeed, one needs to consider the flux line lattice rather than a single flux line. We expect that the elastic properties of the vortex lattice, in particular the elastic moduli c_{11}, c_{44} and c_{66} are also influenced by χ_P. We can then ascribe the peak effect to a vortex lattice melting transition, where the magnetic field at which the transition occurs is strongly suppressed for high values of χ_P due to a softening of the lattice. An elastic theory of the vortex lattice which includes the spin susceptibility of the normal electrons is needed to confirm this conjecture. An explanation in terms of the GFFLO is clearly refuted by our results, with our values of β around 1.2 and of the mean free path of only 3 times the coherence length for $x=0.075$.

4. Summary

In conclusion, we have doped $CeRu_2$ with La. This does not change the intrinsic pinning potential present in the sample, but shifts the field at which the peak opens up to higher values. We attribute this shift to the lowering of the spin susceptibility with increasing La concentration. A qualitative explanation in terms of a melting transition of the vortex lattice seems appropriate, but needs to be confirmed by transport measurements of the flux line lattice and by a pinning theory taking the spin susceptibility into account.

[4] This behavior was actually observed in thin films of $CeRu_2$, where flux lines are indeed individually pinned due to high disorder.

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