

## ANOMALOUS ELECTRODYNAMIC RESPONSE IN THE MIXED-VALENCE SUPERCONDUCTOR CeRu<sub>2</sub>

*N.E.Sluchanko*<sup>1</sup>, *V.V.Glushkov*, *S.V.Demishev*, *N.A.Samarin*, *I.B.Voskoboinikov*,  
*O.D.Chystiakov*<sup>+</sup>, *Y.Bruynseraede*<sup>\*</sup>, *V.V.Moshchalkov*<sup>\*</sup>

*General Physics Institute RAS, 117942 Moscow, Russia*

<sup>+</sup>*Institute of Metallurgy RAS, 117334 Moscow, Russia*

<sup>\*</sup>*Lab. voor Vaste-Stoffysica en Magnetisme, B-3001 Leuven, Belgium*

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We report the first results of microwave magnetoabsorption measurements (35–140 GHz) in the intermediate-valence superconductor CeRu<sub>2</sub>. The anomalous electrodynamic response is found to derive from a transition from a weak to a strong pinning regime in the superconducting mixed state of the unusual metal. The experimental results strongly support the appearance in the CeRu<sub>2</sub> mixed state of a first-order phase transition that may be explained in terms of Fulde – Ferrel – Larkin – Ovchinnikov state formation.

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1. Recently much attention has been drawn to the mixed state of the superconducting C-15 Laves phase compound CeRu<sub>2</sub>, which exhibits an anomalous magnetic response in the vicinity of the upper critical field  $H_{c2}$  at temperatures  $T \leq 0.9T_C$  [1–6]. The unconventional peak effect (PE) is observed for samples of different origin [2], including metallurgically clean samples and high-quality single crystals, contrary to the classical PE [7] studied extensively since the 1960s. The unconventional PE in CeRu<sub>2</sub> is very similar to that found in the heavy-fermion superconductors UPd<sub>2</sub>Al<sub>3</sub> [8] and UPt<sub>3</sub> [9]. However, contrary to the widely discussed unconventional superconducting state (SCS) with a multiple-component order parameter in UPt<sub>3</sub> [10], the SCS in CeRu<sub>2</sub> may be interpreted in terms of a single-component order parameter (*s*-wave pairing) in the framework of the strong-coupling BCS model [2].

The origin of the anomalous peak effect in CeRu<sub>2</sub> should be sought within the nature of the phase transition inside the mixed state. In particular, the Fulde – Ferrell [11] and Larkin – Ovchinnikov [12] models, in which the superconducting order parameter is spatially modulated as a function of magnetic field, can be used to consider the first-order phase transition between a weak and a strong pinning regime. More systematic studies of the peak effect, in particular, by means of direct and microscopic techniques, are needed to reveal which intriguing pinning phenomenon accompanies this transition in the mixed state.

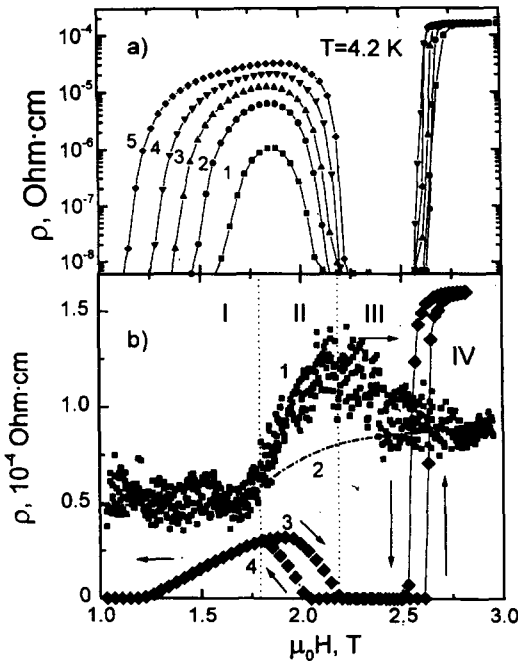
The electrodynamic response of type-II superconductors in the mixed state is strongly affected by vortex dynamics, as has been shown by extensive dc-transport and microwave-frequency studies of both low- and high- $T_C$  superconductors [13–15]. Additionally it has been reported recently [16, 17] that microwave magnetoabsorption (reflection) methods are very convenient experimental tools to study the magnetic phase transitions in Ce-based Kondo-lattice compounds.

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<sup>1</sup>) e-mail: nes@lt.gpi.ru

The aim of the present work is to investigate the features of the unconventional pinning phenomenon. The electrodynamic response has been studied in the frequency range 35–120 GHz in magnetic fields up to 7 T for the normal and superconducting states of CeRu<sub>2</sub>. For example, microwave magnetoabsorption measurements have been performed for frequencies well below the energy-gap value ( $2\Delta \approx 4.0 k_B T_C$  [18]) within the mixed state of the Ce-based superconductor. Simultaneously, direct measurements of the hysteretic peak effect and magnetic susceptibility have been carried out for the same samples of CeRu<sub>2</sub>.

2. Polycrystalline samples of CeRu<sub>2</sub> were synthesized by the arc melting of stoichiometric amounts of the constituent elements 4N (99.99% pure)-Ce and 3N (99.9% pure)-Ru in an argon atmosphere. The sample was annealed in an evacuated quartz tube for two weeks at 900 °C. The longitudinal resistance was measured using dc four-probe techniques with isothermal magnetic field sweeps ( $\mathbf{H} \perp \mathbf{J}$ ) at different rates.



Absorbed power, a.u.

Fig.1. a) Magnetic field dependences of the resistivity  $\rho(H)$  at liquid-helium temperature for different current densities: (1)  $J = 96.5 \text{ A/cm}^2$ , (2)  $J = 115.5 \text{ A/cm}^2$ , (3)  $J = 131.0 \text{ A/cm}^2$ , (4)  $J = 153.9 \text{ A/cm}^2$ , (5)  $J = 192.3 \text{ A/cm}^2$ . b) Magnetic field dependence of the microwave absorption ( $\nu = 45.43 \text{ GHz}$ , curve 1) and resistivity isotherms  $\rho(H, J=192.3 \text{ A/cm}^2)$  (curves 3 and 4) at liquid-helium temperature; I–III — superconducting state for weak and strong pinning regions, IV — normal state

Measurements of the microwave radiation power  $P(H)$  absorbed in the sample were performed on a magneto-optical spectrometer [17] in a magnetic field up to 7 T. The microwave radiation sources used were BWT-generators ( $\nu = 35\text{--}140 \text{ GHz}$ , about 10 mW output). The experimental setup allowed us to carry out magnetoabsorption measurements at temperatures of 1.8–4.4 K. The low-frequency-modulated ( $f = 23 \text{ Hz}$ ) radiation was transmitted by a waveguide into a cryostat containing a vertical magnet. A combination of waveguide accessories and movable quasi-optical teflon lenses was used to focus microwave radiation on the surface of sample. The polycrystalline CeRu<sub>2</sub> sample was mounted on a thin silver diaphragm located on the cold finger. A miniature carbon thermometer for detecting low-frequency-modulated microwave absorption was glued on the opposite (“dark”) surface of sample for better thermal contact. To avoid the influence of heat-release effects, different rates of magnetic field sweeps were used.

3. The family of resistivity  $\rho(H)$  isotherms is shown in Fig.1a for liquid-helium temperature and different current densities  $J$  in the interval 0–200 A/cm<sup>2</sup>. The increase of  $J$  is accompanied by a dramatic elevation of the  $\rho(H)$  amplitude in intermediate magnetic fields  $H < H_{c2}$ , and additionally the resistivity anomaly spreads over a wide interval of magnetic fields (Fig.1a). Contrary to the classical PE phenomenon there is no “peak effect region” for  $H < 0.7$  T, which rules out the scaling-law behaviour found empirically for the classical PE [7]. Moreover, the state I (Fig.1b) is much more reversible than in the classical cases, for which the critical current remains everywhere at least a few percent of its value at the peak height. The electrical resistivity isotherms Fig.1a,b are similar to the results obtained in [19, 20] for thin films and polycrystalline samples of CeRu<sub>2</sub>.

The hysteresis in  $\rho(H)$  (Fig.1b, region II on curves 3–4) is accompanied by the appearance of a substantial nonmonotonic microwave magnetoabsorption signal in the mixed state of CeRu<sub>2</sub>. The typical field dependence of the microwave absorption at  $T = 4.2$  K is presented in Fig.1b for  $\nu \approx 45.4$  GHz (curve 1) together with magnetoresistance data  $\rho(H, T=4.2$  K) for  $J = 192.3$  A/cm<sup>2</sup> (curves 2,3). Note that hysteresis (region II in Fig.1b) is always present at the onset of the anomalous pinning region, and hence the related nonmonotonic magnetoabsorption in the CeRu<sub>2</sub> mixed state (Fig.1b, intervals II–III of curve 1) can be regarded as a characteristic feature of a strong pinning regime below  $H_{c2}(T)$ .

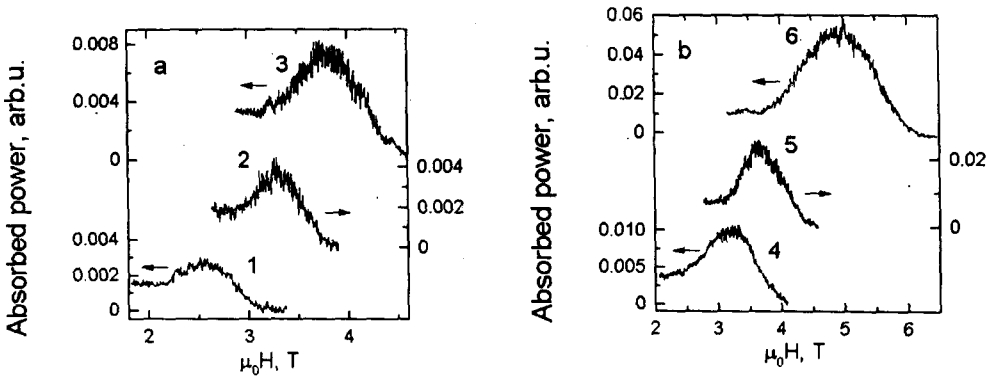


Fig.2. Microwave magnetoabsorption in CeRu<sub>2</sub> in the mixed state for  $\nu = 37.2$  GHz (a) and  $\nu = 99.4$  GHz (b) at different temperatures: (1) 4.35 K, (2) 3.66 K, (3) 3.23 K, (4) 3.68 K, (5) 3.26 K, (6) 2.19 K

In Fig.2 we present the family of magnetoabsorption  $P(H)$  curves obtained at different temperatures between 1.8–4.4 K for microwave frequencies of 37.2 GHz and 99.4 GHz. Note that the magnitude of the SCS  $P(H)$  anomaly increases substantially as the temperature decreases, and at the same time an enlargement of the interval of this mixed-state electrodynamic response is also observed. To summarise the  $P(H)$  data taken at different temperatures, the features of the SCS magnetoabsorption (the initial  $H^*$  and final  $H_{c2}$  values of the  $P(H)$  maxima) are plotted in Fig.3a. The magnetic  $H$ – $T$  phase diagram deduced in the aforementioned manner from our microwave absorption data is presented in Fig.3a. This phase diagram is in good agreement with results published previously [1–6]. In addition, the temperature of the  $P(H)$  maximum is plotted on the  $H$ – $T$  plane (Fig.3a).

It is well established for conventional type-II superconductors that microwave measurements are not hindered by pinning effects [21]. Since the depinning frequency is found

to be in the MHz range for several low- $T_C$  superconductors [22,23], we can expect that for the SCS in CeRu<sub>2</sub> the microwave magnetoabsorption at frequencies 35–120 GHz, well below the energy gap ( $2\Delta \approx 4.0k_B T_C$  [18]), will contain basic information about the power absorption (the time-averaged rate of energy dissipation per unit area) and the penetration depth. However, the penetration depth usually varies monotonically with magnetic field and frequency [19, 24], and it is therefore natural to expect a smooth variation of  $P(H)$  in the vicinity of the transition region (see curve 2 in Fig.1b). Since experiment shows a strongly nonmonotonic behaviour of the magnetoabsorption, the anomalous  $P(H)$  peak in Figs.1,2 can be attributed mostly to the effect of an additional power absorption during the phase transformation in the mixed state.

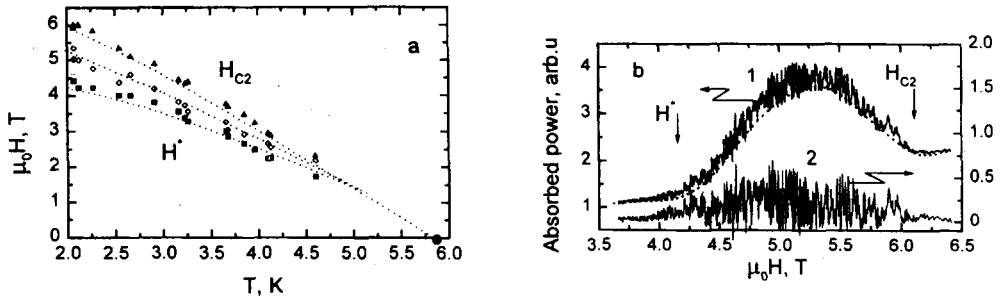


Fig.3. a) Magnetic  $H$ - $T$  phase diagram as deduced from  $P(H, T_0)$  results obtained at different temperatures (1.8–4.4 K) and microwave radiation frequencies (35–120 GHz):  $\bullet$  —  $T_C$  value from the magnetic susceptibility measurements of the same CeRu<sub>2</sub> sample. b) Superconducting mixed-state anomaly of the microwave magnetoabsorption signal  $P(H, T_0=2.12\text{ K})$ ,  $\nu = 37.2\text{ GHz}$  (1) and its “fine structure” (2) (see text)

From this point of view it is very difficult to attribute our microwave magnetoabsorption results (Figs.2,3) to the presence of the microstructure containing slightly different superconducting characteristics, as was recently suggested for CeRu<sub>2</sub> from small-angle neutron scattering investigations [25]. Moreover, our X-ray diffraction measurements, together with the magnetic susceptibility and resistivity data and also preliminary results of Hall coefficient and thermopower experiments, allow us to consider the CeRu<sub>2</sub> samples investigated here as homogeneous superconductors with only a small variance of  $T_C$  (the detailed description of these results will be published elsewhere).

It is very interesting to note that there is an additional “fine structure” of the magnetoabsorption signal inside the interval  $H^* \leq H \leq H_{c2}$ , which can be interpreted in terms of an enhanced noise in the vicinity of the  $P(H)$  maxima (Fig.3b, curves 1, 2). In this situation one cannot rule out the appearance in the  $P(H)$  signal of an additional contribution due to collective pinning effects arising from the presence of the superconductive layered structure consisting of periodic sheets with a vanishing order parameter perpendicular to the applied field [26]. Another possible explanation for the anomalous microwave magnetoabsorption (Figs.2,3) could be the formation of a long-wavelength magnetic order which might also evolve with temperature and field, but at present there is not enough experimental evidence [27] to support this kind of interpretation.

To summarise, we have reported the first results of microwave magnetoabsorption measurements in CeRu<sub>2</sub> in the superconducting mixed state. An anomalous electrodynamic response is found in the frequency range (35–140 GHz), well below the superconducting

gap in this intermediate-valence compound, and can be attributed to a transition from a weak to a strong pinning regime. The experimental results may be explained in terms of a first-order phase transition in the CeRu<sub>2</sub> mixed state, which is not inconsistent with the Fulde – Ferrel – Larkin – Ovchinnikov state formation in this unusual superconductor.

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