

Anomalous magnetization behavior of single-crystalline CeRu₂

Kazuo Kadowaki

*Institute of Materials Science, The University of Tsukuba, Tsukuba, Ibaraki 305, Japan
and National Research Institute for Metals, 1-2-1 Sengen, Tsukuba, Ibaraki 305, Japan*

Hiroyuki Takeya and Kazuto Hirata

National Research Institute for Metals, 1-2-1 Sengen, Tsukuba, Ibaraki 305, Japan

(Received 17 August 1995; revised manuscript received 16 January 1996)

Dc- and ac-magnetic measurements have been performed in the superconducting state of single-crystalline CeRu₂ grown by the Czochralski method in order to understand the anomalous pinning mechanism associated with the peak effect. It is found that with increasing magnetic field the dc-magnetization hysteresis loop closes at a relatively low magnetic field, $\mathbf{B}_{\text{irr}}(T)$, above which it becomes reversible completely. It is also observed that the hysteresis loop abruptly opens up again at $\mathbf{B}_p(T)$ near $\mathbf{B}_{c2}(T)$. Since the onset field of this anomalous peak effect with ascending field $\mathbf{B}_{p,u}$ does not match the ending field with descending field $\mathbf{B}_{p,d}$, this sudden appearance (disappearance) of magnetic hysteresis at the different magnetic field is considered to be the first-order transition. The vortex phase diagram in CeRu₂ is argued in terms of the possible pinning mechanisms in this compound. [S0163-1829(96)01622-0]

I. INTRODUCTION

Recently, there has been a growing interest in the anomalous peak effect observed in heavy fermion compounds such as UPd₂Al₃ (Ref. 1) and UPt₃,² and in some A-15 compounds such as V₃Si (Ref. 3) and Nb₃Sn.⁴ Although the phenomena itself are known to exist in many conventional type-II superconducting materials, the mechanism of the peak effect in most cases is not well understood except for a few exceptional cases. By surveying the peak effect in different materials, one common feature is found: the peak effect occurs only in the pure materials with extremely weak pinning forces. By this reason, the peak effect is observed only in very good single crystals. Since in such materials it is expected that there is a small number of defects working as strong pinning centers, the remaining point defects such as vacancies may play an important role for the occurrence of the peak effect. It is noted that the anomalous peak effect in CeRu₂ described here is not the exception in this sense.

The special interest in the heavy fermion compounds has been raised from the intriguing speculation that the symmetry of the superconducting order parameter in such systems may not be of simple *s*-wave type, but that with higher angular momentum. This speculation has naturally led to a statement that since the superconducting state must be of multiphase nature due to multiple degrees of freedom, unconventional pinning forces may occur due to the anomalous pairing mechanism. The peak effect observed in CeRu₂ also lies in this line, since this compound is known to be a valence fluctuating system with renormalized electronic state similar to the heavy fermion compounds.⁵

A striking similarity in the phenomenon of the peak effect of CeRu₂ has been found in a class of high-*T_c* cuprate superconductors. The typical compounds are the ones with large anisotropy in the superconducting state such as Bi₂Sr₂CaCu₂O_{8+ δ} ,⁶ Tl₂Ba₂CaCu₂O_{8+ δ} ,⁷ RBa₂Cu₄O_{8+ δ} (124 type),⁸ where *R* stands for the rare-earth elements, etc. It

seems that the peak effect observed in these compounds is different from the one observed in most of the other high-*T_c* compounds such as La_{2-*x*}Sr_{*x*}CuO_{4- δ} ,⁹ RBa₂Cu₃O_{7- δ} (123 type),¹⁰ etc. with relatively smaller anisotropy in the superconducting phase. In the case of the 123 type, for example, the peak effect occurs rather gradually with respect to magnetic field, being in sharp contrast with the abrupt occurrence of the peak effect at a certain field strength in the former case. Therefore, the origin of the peak effect in the later case has been thought to be different, for example, from the case of Bi₂Sr₂CaCu₂O_{8+ δ} and has been considered to be the site exchange disorder effect at the Ba and *R* ions as well as the effect of the oxygen nonstoichiometry.¹⁰ For the case of Bi₂Sr₂CaCu₂O_{8+ δ} in particular, it was shown by the neutron-diffraction study¹¹ and the recent muon-spin rotation experiments¹² that the peak effect is driven by the three-dimensional–two-dimensional (3D–2D) transition of the vortex lattice due to the dominant 2D collective-pinning interaction above the crossover field of about 0.05–0.1 T.

Here, we present the results of our recent experimental study on the single-crystalline CeRu₂ by means of dc- and ac-magnetization measurements. The abrupt appearance of the peak effect in an unconventional fashion in this system suggests that the vortex state may undergo the first-order phase transition at \mathbf{B}_p . Such an anomalous occurrence of the peak effect and the vortex phase diagram in CeRu₂ is argued in terms of the possible mechanisms of the first-order transition in the vortex state.

II. EXPERIMENTS

The polycrystalline samples of CeRu₂ were prepared by arc-melting of a stoichiometric amount of the constituent elements under an argon atmosphere. The samples were then set in the water-cooled cold crucible in the tetra-arc single-

crystal growing furnace. A piece of single-crystalline CeRu_2 was attached to the upper shaft as a seed crystal and the Czochralski method was used to grow single crystals. The growth rate was 3–5 mm/h. The size of the single crystal obtained was about 6 mm in diameter and 40 mm in length. Experimental details of the single-crystal growth will be described elsewhere.¹³

The crystallinity of the single crystal was examined by the x-ray back Laue method and by the powder-diffraction technique. Only sharp spots assigned to the cubic Laves phase of CeRu_2 were observed in the back Laue picture, and no other phases in the powdered single crystals were identified in the x-ray powder-diffraction patterns. Moreover, the microstructural study carried out by scanning electron microscopy, energy dispersive x-ray analysis, and electron probe microanalysis (EPMA) showed a clearly good single phase single crystal and is confirmed that the single crystal is sufficiently in high quality.

The sample for the magnetization measurements was cut from the bulk single crystal into a size of 1.1 mm \times 1.0 mm \times 4.0 mm. The superconducting transition temperature T_c was determined by dc magnetization in 0.1 mT to be 6.10 K. The residual resistivity is also measured and was 2–3 $\mu\Omega$ cm at 4.2 K. This reconfirms again that this single crystal is high quality.

The dc- and ac-magnetization measurements were performed by a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-5S) up to 5 T applied parallel to the [001] direction. The ac magnetization at a fundamental frequency of 110 Hz was measured with ac fields of 0.01, 0.1, and 0.38 mT under a dc field up to 5 T. Both ac- and dc-magnetic fields in this case were applied along the [001] direction of CeRu_2 .

III. RESULTS AND DISCUSSIONS

A. dc magnetization as a function of magnetic field

An example of the hysteresis loops in magnetization of CeRu_2 are shown in Fig. 1 measured at various temperatures. The magnetization curves behave as a typical conventional type-II superconductors except for the additional features described below.

Firstly, the magnetization becomes reversible at a relatively low field B_{irr} above the initial hysteretic region, which is located just above $B_{c1} = 0.03\text{--}0.035$ T. As a result, the reversible magnetization region extends to a major part of the mixed state. It is surprising that this reversibility of magnetization was also observed even in rather impure polycrystalline samples containing considerable amounts of foreign phases, which is, in the conventional sense, expected to act as strong pinning centers. Therefore, this unusual weak pinning behavior naturally leads to a speculation that there may exist an intrinsic weak pinning mechanism, which may cause the anomalous pinning behavior in this material.

Secondly, with further increasing magnetic field, the reversible magnetization suddenly becomes irreversible (hysteretic) at around B_p (which will be redefined to be $B_{p,u}$ for ascending field and $B_{p,d}$ for descending field) and it finally closes near B_{c2} . The overview of this anomalous magnetization behavior (peak effect in general) is presented in Fig. 1.

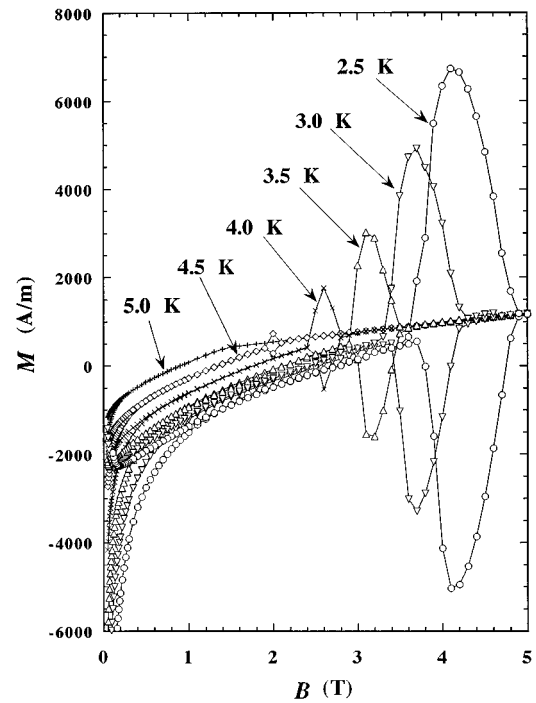


FIG. 1. A set of hysteresis loops in dc magnetization of single-crystalline CeRu_2 at temperatures of 2.5, 3.0, 3.5, 4.0, 4.5, and 5.0 K. The magnetic field is applied to the [001] axis.

In Fig. 2, only the hysteresis loops in magnetization at various temperatures are shown in detail. The following characteristics can be drawn from a closer look at these results:

- (1) The hysteresis loop in magnetization (the peak effect) becomes larger in size as temperature is lowered.

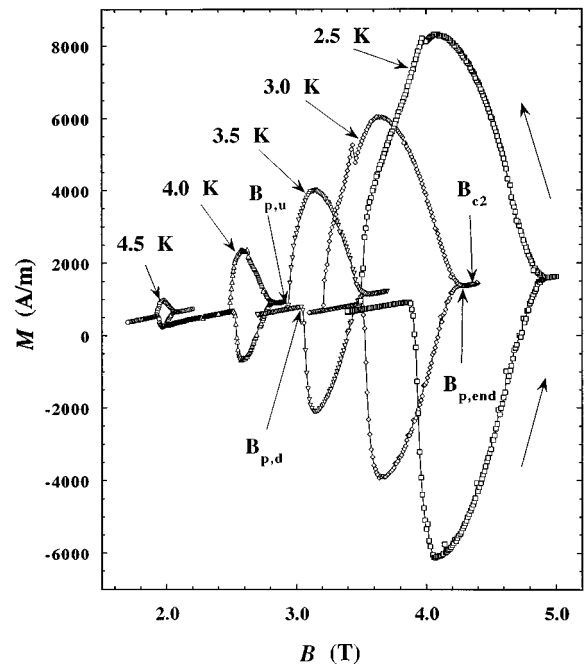


FIG. 2. A set of hysteresis loops in dc magnetization of single-crystalline CeRu_2 only in the peak effect region at temperatures of 2.5, 3.0, 3.5, 4.0, and 4.5 K. The scale in Fig. 1 is expanded. The magnetic field is applied to the [001] axis.

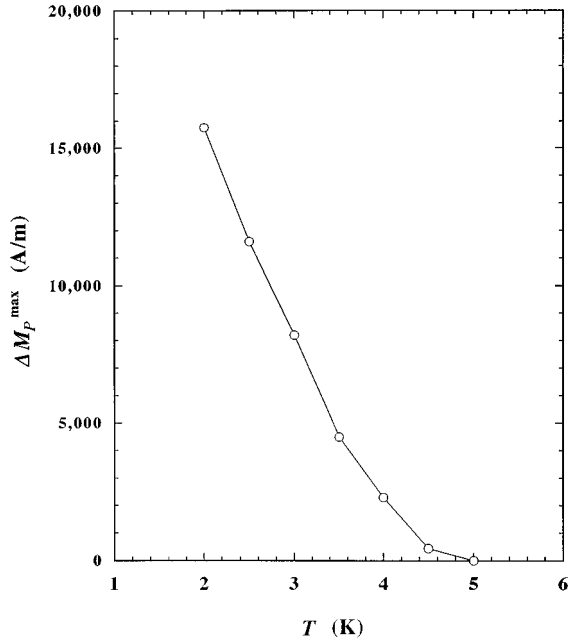


FIG. 3. Temperature dependence of the maximum difference in magnetization $\Delta M = M^+ - M^-$ in single crystalline CeRu_2 at the peak effect.

(2) The onset field of the peak effect at \mathbf{B}_p is very abrupt and it occurs at a slightly higher field $\mathbf{B}_{p,u}$ with ascending field than that with descending field $\mathbf{B}_{p,d}$, as clearly seen in Fig. 2.

(3) When the magnetic field is reversed in the middle of the peak effect, the magnetization follows exactly the profile of the whole hysteresis loop (not shown here).

(4) The closing point of magnetization in the peak effect $\mathbf{B}_{p,\text{end}}$ does not coincide with \mathbf{B}_{c2} . As is seen in Fig. 2, $\mathbf{B}_{p,\text{end}}$ is somewhat lower than \mathbf{B}_{c2} , which is determined by the resistivity measurements.

The results shown in (1) can be understood as a common feature of pinning phenomena in superconductors. This is simply a manifestation of the temperature dependence of the maximum hysteresis $\Delta M_p^{\text{max}} = M^+ - M^-$ as shown in Fig. 3, which reflects the temperature dependence of the superconducting order parameter relevant for the pinning. From the magnetization data as a function of field the macroscopic pinning force F_p can be obtained by a formula of $F_p = \mathbf{J}_c \times \mathbf{B}$. This is shown in Fig. 4. In the inset of Fig. 4, the normalized macroscopic pinning force F_p/F_p^{max} is plotted as a function of normalized field $b = B/B_{c2}$. As is seen in Fig. 4, the normalized pinning force does not scale into the universal curve by the scaling law, which is commonly observed in the peak effect in conventional superconductors.³ It is evident from Fig. 4 that the reason for this is that the onset does not obey the scaling law.

The fact shown in (2) implies an important evidence that the occurrence of the peak effect may be the first-order transition in the sense that the occurrence is very sharp and hysteretic. In particular, the sharpness is in strong contrast to the peak effect observed in other systems such as Ti added 20% Nb alloys,¹⁴ V_3Si ,³ $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-\delta}$,⁹ $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$,¹⁰ etc., where the occurrence of the peak effect is not sharp but rather continuous as a function of magnetic field. This fact

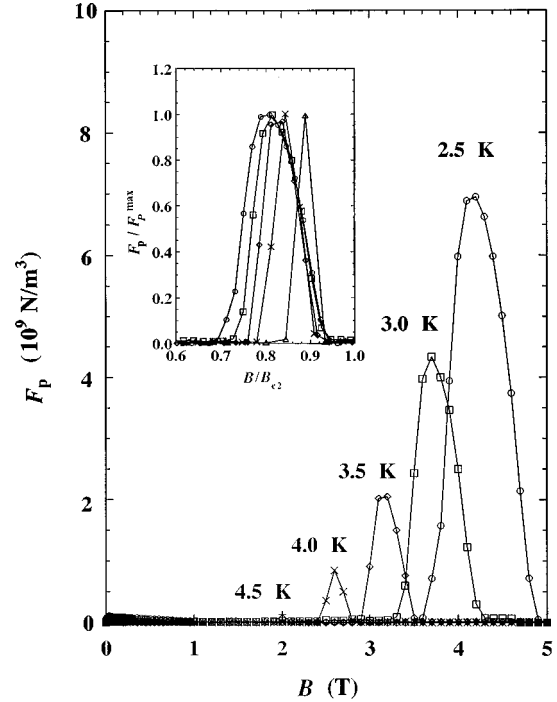


FIG. 4. The macroscopic pinning force density F_p in single-crystalline CeRu_2 as a function of field at various temperatures. The inset shows the normalized pinning force density as a function of reduced magnetic field $b = B/B_{c2}$.

strongly implies that the mechanism of the peak effect in CeRu_2 may differ from that of other peak effects in most of conventional superconductors.

The experimental observation shown in (3) represents evidence that the vortex state in the peak effect region consists of the critical state with full field penetration. The experimental observation given in (4) was initially thought to be an additional anomalous behavior associated with the peak effect. After more careful experimental studies in both ac- and dc-magnetization measurements, however, it turned out that the small difference between $\mathbf{B}_{p,\text{end}}$ and \mathbf{B}_{c2} originates from the relaxation effect in the dc SQUID magnetization measurements. Such a behavior is also seen in the data reported earlier in V_3Si .³

It is noted that the size and the shape of the peak depend rather strongly on the sample as well as the sample shape. The data presented in Figs. 1 and 2 as a example, which show about 14% difference in this case, are taken from the different samples grown by the same technique. This fact implies that the peak may be governed by the subtle energy balance between pinning forces due to the various imperfections inside the crystal and the surface energy barrier which give rise to the inhomogeneous shape-dependent field penetration.

Furthermore, it is interesting to point out that the magnetization at the peak effect region often shows discontinuous jumps, especially at low temperatures. This phenomenon seems to be experimentally reminiscent of the flux jump in conventional hard superconductors, because the position, the frequency, and the sizes of the jump are rather systematic with a fixed sweep rate of the magnetic field as well known in the flux jump.

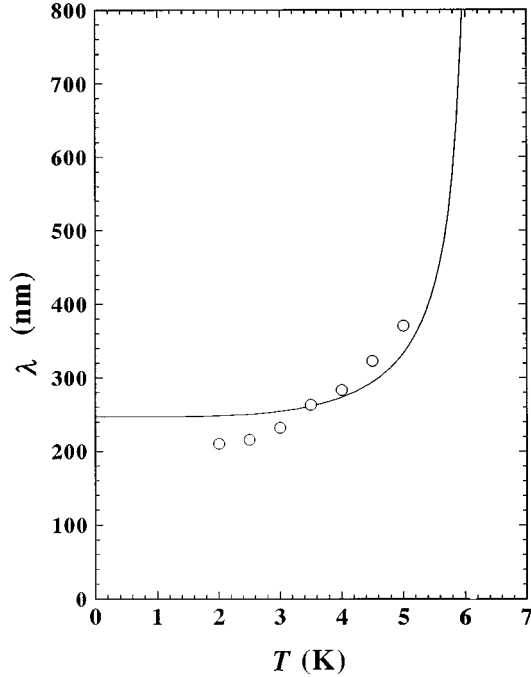


FIG. 5. The temperature dependence of the penetration depth $\lambda(T)$ in single-crystalline CeRu_2 .

Although a considerable variation in the above-mentioned characteristics of the peak effect with the crystal orientation, it is concluded that the intrinsic anisotropy of the peak effect is within a few percent as far as the critical fields are concerned. The reversible magnetization observed in a wide field region shown in Fig. 1 can be analyzed as follows. Firstly, the magnetization in a field region $\mathbf{B}_{c1} \ll \mathbf{B} \ll \mathbf{B}_{c2}$ in the London model can be written as

$$M(B) = -\frac{\phi_0}{32\pi\lambda^2(T)} \ln\left(\eta \frac{B_{c2}}{B}\right), \quad (1)$$

where ϕ_0 is the quantum flux, $\lambda(T)$ is the penetration depth, η is a constant close to unity. By fitting this Eq. (1) to the experimental data, the temperature dependence of λ can be deduced. This result is shown in Fig. 5. The solid curve is a fitted curve to the experimental data by assuming the temperature dependence of $\lambda(T)$ in the two-fluid model:

$$\lambda(T) = \frac{\lambda_0}{\sqrt{1 - (T/T_c)^4}}. \quad (2)$$

From this analysis, $\lambda_0(T=0)$ is obtained to be 2470 \AA .

Furthermore, from the change of the slope of magnetization at \mathbf{B}_{c2} , the Ginzburg-Landau parameter κ_2 can be estimated to be 24, according to the equation of Maki:¹⁵

$$\left(\frac{\partial M}{\partial B}\right)_S - \left(\frac{\partial M}{\partial B}\right)_N = \frac{1}{4\pi\beta} \frac{1}{2\kappa_2^2 - 1}, \quad (3)$$

where $\beta = 1.16$.

B. ac magnetization as a function of magnetic fields

In order to study the dynamical properties of the vortices in this compound, ac measurements were performed under

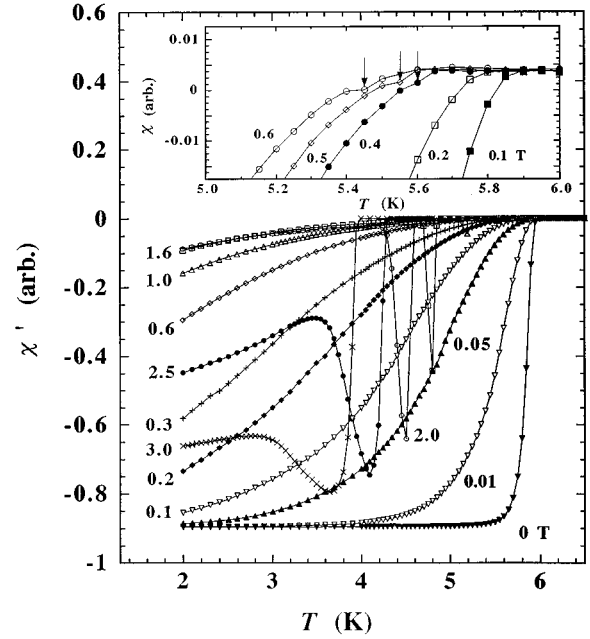


FIG. 6. The set of ac magnetic-susceptibility curves in single-crystalline CeRu_2 as a function of temperature in various magnetic fields. The amplitude and the frequency of the ac magnetic field are 0.38 mT and 110 Hz , respectively. The inset shows the low-field region of the ac susceptibility in expanded scale. The arrows indicate the dip at lower field region, which eventually becomes a sharp dip at higher fields as shown in the main panel.

various dc fields up to 5 T with ac fields of 0.01 , 0.1 , and 0.38 mT . The results are shown in Fig. 6, where the ac field of 0.38 mT is used in this case. Both the real part (χ') and the imaginary part (χ'') of the susceptibility are presented as a function of temperature in a normalized manner. In zero dc field, χ' shows a sharp superconducting transition, whereas the transition becomes broader as the dc field is increased. In addition, a sharp dip in χ' and a peak in χ'' (not shown here) begin to appear in magnetic fields just below T_c . This feature can be more clearly seen in the case of $B = 1$ and 2 T in Fig. 6. At higher fields this sharp peak grows and becomes wider. More detailed behavior only for χ' at lower fields are presented in the inset of Fig. 6 in an expanded scale. The upper critical field, $\mathbf{B}_{c2}(T)$, is in fact determined by the deviating point from the extrapolated normal state one by taking a set of ac susceptibility data similar to Fig. 6 below T_c .

These peculiar experimental results of ac magnetization can be interpreted by a simple model that the pinning causes the hysteresis loss due to an oscillating ac field, which induces macroscopic shielding current in the sample. This shielding current \mathbf{J}_s can be estimated to be $J_s = 3H_{\text{osc}}/4d$ assuming an infinitely long plate with the thickness of $2d$. When the shielding current \mathbf{J}_s becomes higher than $\sim 9 \text{ A/m}^2$, which is induced by $H_{\text{osc}} = 0.38 \text{ mT}$ in this case, this ac magnetization gives the full shielding signal. Therefore, the sharp dip appearing in χ' indicates that the shielding current exceeded at a value determined by the amplitude of the oscillating ac field. The disappearance of the sharp dip in χ' with decreasing temperature as seen in Fig. 6 at low temperatures means that this shielding current became smaller again than the critical value with decreasing temperature.

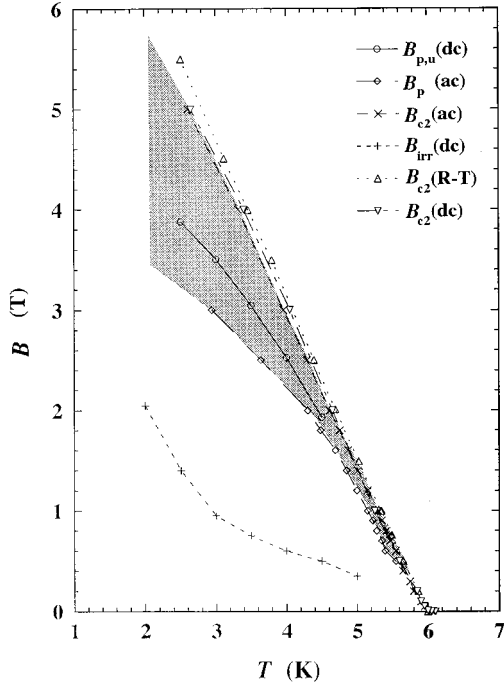


FIG. 7. The magnetic phase diagram of single-crystalline CeRu₂ obtained from the dc magnetization measurements, the ac susceptibility, and the resistive measurements. The shaded area is the region where the peak effect is observed. The boundaries indicated by $\mathbf{B}_{p,u}(\text{dc})$, $\mathbf{B}_p(\text{ac})$, $\mathbf{B}_{c2}(\text{ac})$, $\mathbf{B}_{\text{irr}}(\text{dc})$, $\mathbf{B}_{c2}(R-T)$, and $\mathbf{B}_{c2}(\text{dc})$ stand for the onset field of the peak effect with ascending field, the onset field of the peak effect measured at the dip position in the ac susceptibility, the upper critical field measured by ac susceptibility (defined by the onset of the diamagnetism as a function of temperature), the irreversibility line, the upper critical field determined by the resistivity measurement, and the upper critical field determined by the magnetization measurement, respectively.

This lack of diamagnetic shielding signal can well be ascribed as a lack of pinning force in this material, which corresponds to the reversible magnetization. The broad feature in χ' indicates that the shielding current is not fully established in order to maintain the complete shielding, although the magnetization appears to be reversible within experimental error. This discrepancy can reasonably be ascribed to the different criterion to the critical current density between both experimental techniques used here. This nonzero critical current observed in reversible region in magnetization is also proved by the direct critical current measurement.¹⁶ In general, more stringent criterion can be set in ac-susceptibility measurements than the dc-magnetization measurements.

It is worthwhile mentioning that the anomalous double transition and a possibility of the unusual superconductivity such as the one with the triplet pairing speculated previously,¹⁷ can well be understood by the above-mentioned mechanism caused by the anomalous pinning effect in CeRu₂. However, although the possible scenario of unconventional superconductivity to explain the anomalous peak effect cannot completely be excluded, we believe that the mechanism of the peak effect is an extrinsic effect to superconductivity in case of CeRu₂.

In Fig. 7, the phase diagram deduced by the present experiments is plotted. The shaded area is the region where the

peak effect is observed. It is noted that the boundary \mathbf{B}_p determined by the ac-magnetization measurements is somewhat lower than that determined by the dc SQUID measurements. This is because of the different criterion used for both measurements as explained above. In the ac-magnetization measurements $\mathbf{B}_p(\text{ac})$ is defined by the field indicated by the arrows in Fig. 6. The lack of sharpness of the transition in the ac measurement at $\mathbf{B}_p(\text{ac})$ may be caused by the shielding current flowing inhomogeneously inside the sample due to weak pinning effect existing in the background. In Fig. 7, the irreversibility line \mathbf{B}_{irr} determined by dc-magnetization measurement is also added.

Although the phase boundaries \mathbf{B}_{irr} and \mathbf{B}_p ($\mathbf{B}_{p,u}$ and $\mathbf{B}_{p,d}$) show a considerable sample dependence according to the sample inhomogeneity, the other critical fields \mathbf{B}_{c2} and $\mathbf{B}_{p,\text{end}}$ do not show anisotropy within an accuracy of few percent. Assuming that the upper critical field is governed by the paramagnetic limit (Clongston limit), a value for $\mathbf{B}_{c2}(0)$ can be obtained to be about 11.2 T, which is slightly above the simple extrapolated value of ~ 10.3 T. This indicates that the upper critical field in CeRu₂ is limited by the paramagnetic limit. From the value of $\mathbf{B}_{c2}(0) = 11.2$ T, the coherence length ξ is estimated to be 54 Å. Therefore, the Ginzburg-Landau parameter $\kappa = \lambda/\xi$ is obtained to be ~ 45 .

From the phenomenological Ginzburg-Landau approach \mathbf{B}_{c1} and \mathbf{B}_{c2} can be expressed as

$$B_{c1} = B_c \frac{\ln \kappa_3}{\sqrt{2} \kappa_3} \quad (4)$$

and

$$B_{c2} = \sqrt{2} \kappa_1 B_c, \quad (5)$$

respectively. Therefore, $B_{c2}/B_{c1} = 2\kappa^2/\ln \kappa = 328$, resulting in values for κ and B_c being 23 and 0.35 T, respectively. Here, we used values of $B_{c1} = 0.035$ T and $B_{c2} = 11.5$ T, and it is assumed that $\kappa_1 \sim \kappa_3$. This κ value is in good agreement with the one obtained from Eq. (3), but is nearly twice smaller than the one obtained in the above. The reason for this is not clear but it is certain that the value obtained from the analysis of $\lambda(T)$ shows rather poor agreement with Eq. (2) as shown in Fig. 5 and the $\lambda(T)$ value itself seems to be somewhat too large.

IV. CONCLUDING REMARKS

The experimental results of the anomalous peak effect observed in single-crystalline CeRu₂ were presented and were analyzed with the conventional theory for the type-II superconductors. The sudden appearance (disappearance) of the peak effect at \mathbf{B}_p and the difference of $\mathbf{B}_{p,u}$ and $\mathbf{B}_{p,d}$ strongly suggest that the transition may be a first-order transition.

In the conventional understanding of the peak effect, it is rather difficult to explain such a sharp feature similar to the first-order transition. Possibilities such as the secondary phase inclusions with weak superconductivity identified often in alloys¹⁴ and claimed in $R(=La, Nd, Sm)Ba_2Cu_3O_7$,¹⁸ and the matching effect in some specially arranged correlated pinning centers¹⁹ are definitely excluded, because the samples showing anomalous peak effect including in this

experiment are in very pure form, mostly in high-quality single crystals having neither strong pinning nor correlated pinning centers.

In 1969, Pippard²⁰ proposed a simple mechanism which may be relevant for explaining the peak effect observed in Nb.²¹ The essence of his theory is that the rigidity of the flux-line lattice obeys the quadratic form $[C_{66} \propto B_{c2}(1-b)^2]$ and falls to zero as a function of magnetic field near B_{c2} , whereas the individual pinning force decrease linearly with magnetic field $[f_p \propto b(1-b)]$. Therefore, it is expected that the crossover of the free energy takes place near B_{c2} . Moreover, it was shown that the peak value in J_c may be strengthened by more than an order of magnitude. According to his model, such a transition can, in principle, be very sharp, as sharp as the phase transition. To our knowledge of the peak effect so far studied experimentally in the past, on the contrary to this theoretical prediction, none of them has such a sharp feature except the present CeRu₂. It is still not well understood why only CeRu₂ shows such a sharp transition and it certainly remains as an unsolved question.

According to Larkin and Ovchinnikov,²² it is expected within the collective-pinning theory that there may exist softening of the shear modulus C_{66} due to the renormalization effect, leading to the enhancement of the critical current. This phenomenon was actually confirmed by Kes and Tsuei²³ in the amorphous thin films of Nb₃Ge and Nb₃Si. In these cases, however, the material has an amorphous form and has extremely weak pinning forces with high density. Although the physical conditions in the amorphous materials and the single-crystalline CeRu₂ are completely different, it is intriguing to compare the similarity in both cases from the point of view of weak pinning behavior. Nevertheless, it is again difficult to explain the sharp feature observed in the present experiment within this view.

The unusual enhancement of the pinning force in this system has been thought to be related to the extremely small pinning force in the reversible region. From the similar phenomenon observed in the heavy fermion compounds such as UPd₂Al₃,¹ UPt₃,² etc., a speculation has been made that the large enhanced paramagnetic susceptibility may play a key

role for the occurrence of the peak effect. For the heavy fermion compounds such as UPd₂Al₃ and UNi₂Al₃, it is true that the susceptibility is largely enhanced: for example, it is about 1.3×10^{-3} (SI) for UPd₂Al₃ at low temperatures.^{24,25} Contrary to this, the susceptibility of CeRu₂ is not largely enhanced, but is only 2.6×10^{-4} (SI), which is in good agreement with the previous reports²⁶ and is also in a comparable order with the superconducting transition metals such as V ($=3.6 \times 10^{-4}$ in SI unit), Nb ($=2.36 \times 10^{-4}$), Ta ($=1.6 \times 10^{-4}$), etc. Therefore, these experimental facts certainly pose a question about whether the enhanced paramagnetism is playing an essential role for the occurrence of the peak effect presented here. On the other hand, it is favorable to the weakening of the pinning energy due to the fact that the enhanced paramagnetism reduces the elemental pinning energy, $E_{\text{core,super}} = \pi \xi^2 (B_{c2}/8\pi) \sim 4.5 \times 10^{-12}$ J/m by $E_{\text{core,paramagnetism}} = \pi \xi^2 \chi_{\text{spin}} h^2/2 \sim 1.3 \times 10^{-12}$ J/m, where χ_{spin} is the Pauli paramagnetic susceptibility, and h is the magnetic field inside the vortex core. This effect is about $\frac{1}{3}$ of the total core energy of the vortex in CeRu₂ and could certainly be a major reason to realize the anomalously weak pinning phenomenon in $B_{c1} \ll B \ll B_p$ in CeRu₂. In the same token, it is not clear whether or not the occurrence of the speculated superconducting phase proposed by Fulde-Ferrell²⁷ and Ovchinnikov-Larkin,²⁸ which predict the first-order transition in some cases, is a real cause of the peak effect, although a connection between them has been argued.^{26,29} More systematic study of the peak effect, in particular, by means of direct and microscopic techniques, is needed in order to reveal such an intriguing pinning phenomenon which seems to accompany the first-order transition in the vortex state.

ACKNOWLEDGMENTS

The authors would like to thank Professor M. Tachiki and his collaborators, and Professor F. Steglich and his collaborators for their stimulating suggestions and discussions. This work is supported by the Interdisciplinary Basic Research Program (Shosai Kisokenkyu) under the Science and Technology Agency, Japan.

¹K. Gloos, R. Modler, H. Shimanski, C. D. Bredl, C. Geibel, F. Steglich, A. I. Buzdin, N. Sato, and T. Komatsubara, Phys. Rev. Lett. **70**, 501 (1993).

²T. Sakakibara *et al.* (unpublished).

³M. Ishino, T. Kobayashi, N. Toyota, T. Fukase, and Y. Muto, Phys. Rev. B **38**, 4457 (1988).

⁴A. J. Arko, D. H. Lowndes, F. A. Muller, L. W. Roeland, J. Wolfart, A. T. van Kessel, H. W. Myron, F. M. Mueller, and G. W. Webb, Phys. Rev. Lett. **40**, 1590 (1978), and the references therein.

⁵K. Kadowaki, H. Takeya, and K. Hirata, (unpublished).

⁶K. Kadowaki and T. Mochiku, Physica C **195**, 127 (1992).

⁷V. N. Kopylov, A. E. Koshelev, I. F. Schegolev, and T. G. Togonidze, Physica C **170**, 291 (1990).

⁸M. Xu, D. K. Finnemore, G. W. Crabtree, V. M. Vinokur, B. Dabrowski, D. G. Hinks, and K. Zhang, Phys. Rev. B **48**, 10 630 (1993).

⁹T. Kobayashi, Y. Nakayama, K. Kishio, T. Kimura, K. Kitazawa, and K. Yamafuji, Appl. Phys. Lett. **62**, 1830 (1993).

¹⁰J. L. Vargas and D. C. Larbalestier, Appl. Phys. Lett. **60**, 1741 (1992).

¹¹R. Cubitt, E. M. Forgan, G. Yang, S. L. Lee, D. Mck. Paul, H. A. Mook, M. Yethiraj, P. H. Kes, T. W. Li, A. A. Menovsky, Z. Tarnawsky, and K. Mortensen, Nature (London) **365**, 407 (1993).

¹²S. L. Lee, P. Zimmermann, H. Keller, M. Warden, I. M. Savić, R. Schauwecker, D. Zech, R. Cubitt, E. M. Forgan, P. H. Kes, T. W. Li, A. A. Menovsky, and Z. Tarnawski, Phys. Rev. Lett. **71**, 3862 (1993).

¹³H. Takeya and K. Kadowaki, J. Cryst. Growth (to be published).

¹⁴C. Baker and J. Sutton, Philos. Mag. **19**, 1223 (1969).

¹⁵K. Maki, Physics **1**, 21 (1964); **1**, 127 (1964).

¹⁶K. Kadowaki and H. Takeya (unpublished).

¹⁷K. Yagasaki, M. Hedou, and T. Nakama, J. Phys. Soc. Jpn. **62**, 3825 (1993).

- ¹⁸M. Murakami, Sang-Im Yoo, T. Higuchi, T. Oyama, N. Sakai, and S. Tanaka (unpublished).
- ¹⁹H. Raffy, J. C. Renard, and E. Guyon, *Solid State Commun.* **11**, 1679 (1972).
- ²⁰A. B. Pippard, *Philos. Mag.* **19**, 217 (1969).
- ²¹C. S. Tedmon, Jr., R. M. Rose, and J. Wulff, *J. Appl. Phys.* **36**, 829 (1965).
- ²²A. I. Larkin and Yu. N. Ovchinnikov, *J. Low Temp. Phys.* **34**, 409 (1979).
- ²³P. H. Kes and C. C. Tsuei, *Phys. Rev. Lett.* **47**, 1930 (1981).
- ²⁴C. Geibel *et al.*, *Z. Phys. B* **83**, 305 (1991).
- ²⁵C. Geibel *et al.*, *Z. Phys. B* **84**, 1 (1991).
- ²⁶A. D. Huxley, C. Paulsen, O. Laborde, J. L. Tholence, D. Sanchez, A. Junod, and R. Calemuczuk, *J. Phys. Condens. Matter* **5**, 7709 (1993).
- ²⁷P. Fulde and R. A. Ferrell, *Phys. Rev.* **135**, 550 (1964).
- ²⁸A. I. Larkin and Yu. N. Ovchinnikov, *Sov. Phys. JETP* **20**, 762 (1965).
- ²⁹M. Tachiki, S. Takahashi, P. Gegenwart, M. Weiden, M. Lang, C. Geibel, F. Steglich, R. Modler, C. Paulsen, and Y. Onuki, *Z. Phys. B* (to be published).