Critical current densities in superconducting materials

P CHADDAH

Cryogenics and Superconductivity Section, Centre for Advanced Technology, Indore 452 013, India e-mail: chaddah@cat.ernet.in

Abstract. We discuss recent research in the area of critical current densities (J_C) in superconductors. This shall cover recent work on newly discovered superconductors, as well as on the magnetic-field dependence of J_C .

Keywords. Critical current density; superconducting materials; magnetic-field dependence.

1. Introduction

The electrical resistance of a superconducting material drops to zero below its critical temperature T_c , and the Joule heating given by $i^2 R$ drops to zero. The largest commercial applications of superconductors have been based on this ability to carry current without resistance, and have been in the form of superconducting magnets producing high magnetic fields. Resistive electromagnets produce large local heating due to $i^2 R$ dissipation, the current densities flowing in them cannot be raised beyond a point (about 1000 amps/cm²), and they are commonly used to produce magnetic fields up to 2 tesla only. Kammerleigh Onnes had tried developing superconducting magnets in 1913 itself, but it was soon discovered that most elemental superconductors lose this property at modest magnetic fields (H_C) of around 0.1 tesla. These materials are known as type I superconductors, and are perfect diamagnets (or exist in the Meissner state) when they are superconducting. It was realised in the 1950s that most alloys and compound superconductors belong to a second class of superconductors called Type II superconductors. In magnetic field vs temperature space, the phase diagram of this class of superconductors is comprised of two distinct superconducting regions. There is an upper critical field line $H_{C2}(T)$ above which the material is in its normal resistive state, and there is a lower critical field line $H_{C1}(T)$ below which the material has zero resistance and is also a perfect diamagnet in that there is no magnetic flux penetrating the bulk of the superconductor. These are depicted schematically in figure 1. The region between these two lines corresponds to the superconductor being in the mixed state. In this state the material has zero electrical resistance but is not a perfect diamagnet. In the region between the $H_{C1}(T)$ and the $H_{C2}(T)$ lines, magnetic flux penetrates the superconductor in the form of long vortices, with each vortex carrying an identical quantum of flux. These vortices are regions of normal material in that they do not have any Cooper pairs, and thus have a higher energy than the surrounding superconducting region. They carry identical cylindrical current shells on their periphery, repel each other, and form a triangular lattice in a uniform background. It is this mixed state, of type II supercon-





Figure 1. The schematic magnetisation behaviour of type I and type II superconductors is shown. In the Meissner state they behave as perfect diamagnets, and so magnetic flux penetrates the bulk of the sample. The mixed state of type II superconductors has zero resistivity but allows partial flux penetration, in the form of quantised vortices.

ductors with large values of H_{C2} , that is used for carrying current without resistance. Superconducting magnets, accordingly, use type II superconductors with large H_{C2} and high T_C .

In addition to the two limiting parameters T_C and H_{C2} , which are intrinsic characteristics of a superconducting material, the zero resistance property of the superconducting state is also lost if the material carries a current density J larger than a critical value J_C . As current densities are raised to higher than J_C the material does not go suddenly from the superconducting state to the normal resistive state, but develops a 'flux-flow resistivity' that rises with increasing J. To understand the origin of J_C we now look in some detail at the mixed state that exists for magnetic fields between $H_{C1}(T)$ and $H_{C2}(T)$.

As mentioned earlier, in the mixed state the magnetic flux penetrates the sample in the form of vortices carrying unit flux quanta. These vortices repel one another and minimise their free energy by forming a triangular lattice. Since the vortex density is uniform, the repulsive interactions between any vortex and all its neighbours cancels out. It follows from Maxwell's equation that since Curl $\mathbf{B} = 0$ when the vortices form a uniform triangular lattice, no macroscopic currents flow within the superconductor. We now consider what happens when current is passed through a superconductor in its mixed state. A nonzero J requires a nonvanishing Curl **B**, and this requires a gradient in the vortex density. The repulsive interaction with vortices on either side will no longer cancel out, since the density gradient implies unequal distances between vortices (Wilson 1983). Each vortex thus experiences a net force which is proportional to both the density of vortices and to its gradient, i.e. to both **B** and **J**. This driving force can be derived rigorously, and one obtains $\mathbf{F} = \mathbf{B} \times \text{Curl } \mathbf{B} = \mathbf{B} \times \mathbf{J}$. This force on each vortex, which is equivalent to a Lorentz force, will cause the vortices to move. The vortex motion produces an electric field parallel to **J**, thus causing a resistance, and this is called the flux-flow resistance. The resistance is much smaller than the normalstate resistance, but the material is no longer having infinite conductivity (Campbell & Evetts 1972; Wilson 1983).

We have argued that the existence of a macroscopic current in the mixed state of a superconductor results in a resistance associated with the motion of vortices. It is necessary to prevent this vortex motion if current is to be passed without dissipation. This is achieved by 'pinning' vortices at impurity sites. These impurity regions can be point-defects or linedefects, and they provide optimum pinning when their dimensions are close to the coherence length of the Cooper pairs in the superconductor. (This varies from tens of nm for conventional superconductors to about 1 nm for high T_C materials. The nature of the pinning sites can therefore vary.) To understand how pinning is achieved, consider an impurity region that remains normal when the parent material is superconducting. When a vortex passes through this impurity, the total energy is lower than when the vortex lies just outside the impurity. This is because there are more Cooper pairs in the sample in the former case, while in the latter case the condensation energy is missing both in the vortex and in the impurity. Detailed calculations of the pinning potential of such impurities are commonly done (Matsushita & Ekin 1994), but are outside the scope of this paper. We just point out that the macroscopic pinning force on the entire system of vortices in a sample is the resultant of various pinning forces between individual vortices and impurity sites, and the interaction between vortices themselves. The process of calculating the resultant is quite complicated, and has not yet reached a level where it has determined the actual fabrication procedures of practical superconductors. It is clear, however, that the pinning force rises with rise in number of individual pinning (or impurity sites, but that the pinning force is lowered due to the rigidity of the vortex lattice which hinders distortions necessary for various vortices to pass through the random pinning sites. This has two implications of interest to us. First is that the rigidity of the vortex lattice falls at fields close to $H_{C2}(T)$, and the macroscopic pinning force then rises suddenly. This sharp rise in the pinning force results in the 'peak effect' in J_C (Matsushita & Ekin 1994, sec $3 \cdot 3, 3 \cdot 6$) which has been the subject of many detailed experimental studies, and which we shall address. Second is the idea of APC (artificial pinning centres) approach in which the pinning centres are not located randomly, but their distances are optimised for a particular magnetic field. The pinning centre separation matches the vortex separation for fields close to this magnetic field. The vortex lattice is thus minimally distorted when vortices are pinned at this field value, and the net pinning force remains large.

The pinning force prevents the motion of vortices until the Lorentz force $(\mathbf{J} \times \mathbf{B})$ exceeds the pinning force \mathbf{F}_P . This defines a critical current density $J_C = F_P/B$ below which transport current is carried without any resistance, and above which flux-flow resistivity sets in. As stated earlier, F_P depends on various factors including the field B, and J_C can have a complicated field and temperature dependence. The zero resistance property of a superconductor exists within an operating volume in the field-temperature-current density space, enclosed by the J_C , H_{C2} , and T_c lines. This operating volume is shown schematically in figure 2.

In the next section we describe how J_C can be measured. This is especially important because, unlike T_C and H_{C2} , it is not an intrinsic property of a material but is dictated also by material processing. We shall then discuss some recent research results on recently discovered superconductors that hold the promise for applications. We shall then discuss the peak effect in J_C , and present unusual features observed over the last few years. We shall conclude with our interpretation of these unusual history effects.

2. Measurement of J_C

The direct measurement of J_C involves the measurement of a V-I curve where the current at which a measurable voltage first appears would be considered as the technologically relevant



Figure 2. The operating volume of a superconducting material, where it has zero resistivity, is enclosed by the T_C , H_{C2} and J_C lines. The figure shows these lines for NbTi and Nb₃Sn, the two superconducting materials in commercial use.

 J_C . This somewhat abstract criterion has been quantified and standardised taking into account voltage levels that are comfortably measurable (10⁻⁶ volts), and current densities that are usually desired (around 10⁵ amps/cm²). The standard criterion states that the (flux-flow) resistivity is 10⁻¹² ohm.cm at $J = J_C$. Measurements at this level of accuracy are essential before any material (or any fabrication process) can be considered for commercial current-carrying applications. These measurements require reasonable quantities of sample, fabricated in forms such that current contacts can be easily made.

 J_C can also be inferred from magnetisation vs field (M-H) measurements because any change in external field sets up persistent shielding currents. (How long these currents persist depends on the resistivity as well as the inductance seen by the current loops, and the time scale of measurement of M can be related to the resistivity.) Bean's critical state model (Bean 1964) is used to infer J_C from the hysteresis in the M-H curves. The field dependence of J_C can also be obtained, with some provisos (Chaddah *et al* 1989), from M-H curves. This method is very popular at the research level and is used on small quantities of new materials. The inferred J_C does not, however, usually reflect the weak links in the sample because the currents passing through them do not persist for long times.

3. J_C in technologically relevant superconductors

NbTi and Nb₃Sn are the superconductors in commercial use today, and their $J_C-T_C-H_{C2}$ curves are shown in figure 2. Of these, NbTi is used in over 80% of the current-carrying applications. The fabrication process of multistrand wires involves not just alloy (for NbTi) or compound (for Nb₃Sn) formation, but also coldwork during wire drawing. These processes create impurity pinning sites, and are frequently discussed (Wilson 1983; Asner 1989; Chaddah *et al* 1989). While α -titanium precipitates are the pinning centres usually formed in NbTi during alloy formation, there have recently been efforts to replace these by 'artificial' pinning centres whose individual dimensions and neighbouring distances can be varied in a controlled way. A 'repulsive' pinning is provided by thin layers of niobium that have been introduced artificially, and which become superconducting due to the proximity effect. These superconductors can then be optimised to have large J_C at particular fields because the vortex

lattice-constant would then match with the inter-pinning-site separation, thereby maximising pin-to-vortex overlap without distorting the vortex lattice. This idea has met with encouraging results (Asner 1989).

Amongst the new superconductors with a promise for applications, we shall consider two, viz. YBa₂Cu₃O₇ (and other rare-earth-based 123 oxides) and MgB₂. Clearly, this does not exhaust the list of promising superconductors but only reflects the present author's interests. The former is a representative of the high T_C oxide superconductors which can be cooled to the superconducting state with liquid nitrogen, but whose commercial exploitation has been impeded both by their ceramic nature and by the fact that their J_C decreases sharply at moderate fields. The second represents a metallurgically friendlier compound, with a T_C (39 K) well above that of conventional superconductors, but well below what the cuprate superconductors have now got us used to! We note that the potential for applications will be realised only if J_C is close to 10^5 amps/cm² at temperatures of 20 K (for MgB₂) or 77 K (for YBa₂Cu₃O₇).

Fifteen years have passed since the discovery of high T_C superconductors. Long lengths of wires of Bi-based cuprate materials are being made, but their utilisation is restricted to temperatures around and below 20 K. The values of J_C are low, but their high H_{C2} allows them to be used as high-field inserts in hybrid magnets. They cannot be used close to their T_C (or with liquid nitrogen) because the J_C drops rapidly at modest fields. This limitation is believed to be intrinsic to their layered structure, and to phase transitions in their vortex structure. High T_C superconductors of the YBa₂Cu₃O₇ family (hereafter referred to as YBCO), on the other hand, do not have intrinsic imitations at 77 K. Though long length wires of these materials (with high values of J_C) have not been made, a lot of research effort is going on as outlined below. The coherence length of these materials are small, and defects of dimensions around 1 nm are naturally present in these materials (these include oxygen vacancies and twin boundaries). It is for this reason that it is very difficult to obtain YBCO samples that show reversible magnetic behaviour. Improving J_C by introducing additional defects has been difficult, and the successful attempts include (a) introducing a fine dispersion of an insulating phase (called Y-122 phase); (b) introducing columnar defects by heavy-ion irradiation; and (c) control of oxygen stoichiometry. Of these, (a) introduces non-superconducting particles of dimensions (few tens of nm) much larger than the coherence length. They do act as pinning centres in low fields, but J_C degrades rapidly with increasing field. The best values reported at 77 K are 4×10^4 A/cm² at zero field, 1×10^4 A/cm² at 2 tesla, and dropping to below 10^3 A/cm² at 4 tesla. Procedure (b) introduces columnar defects of atomic dimensions, but the technique is not suitable for large scale production and has not been pursued for applications. The procedure of controlling oxygen stoichiometry is the most promising method, and was introduced by Larbalastier's group in 1990 (Daeumling et al 1990). It was argued that small oxygen deficient regions exist in bulk samples, and these regions have degraded superconducting properties including low H_{C2} . They observed a large peak effect in M–H curves (and thus inferred J_C) in single crystal samples at 70 K, with the peak occurring at around 5 T and the J_C being larger than zero fields J_C ! They attributed this to the oxygen-deficient regions becoming normal at these fields because the H_{C2} of these regions had been exceeded, and then acting as very efficient pinning centres. (It may be noted that this origin of the peak effect is very different from the softening of the lattice discussed in the introduction. This is an active area of research that we shall discuss in the next section. This concept of Daeumling et al (1990) has been applied by Murakami et al (2000) in the oxygen-controlled-melt growth (OGMG) technique of bulk processing to obtain large peak effects in Nd-, Sm-, Eu- and Gd-based 123 superconductors. This peak effect is also believed to be due to magnetic field induced pinning

centres. The peak position depends on the oxygen partial pressure during processing and also the rare-earth elements in the compounds. Typically for Nd-123 at 77 K, J_C values obtained are 2×10^4 /cm² at 0 T, 4×10^4 A/cm² at 2 T and 1×10^4 A/cm² at 4 T. Further research is continuing in numerous labs. And target values of 10^5 A/cm² at 77 K and 3 T are talked about, holding the hope of commercially viable SC magnets at 77 K. There is, however, no satisfactory technique for wire production for the Y-123 family and various techniques for fabrication of thick films are being developed.

Superconductivity in MgB₂ at 39 K was discovered less than a year back, and this material has been taken as a possible new low-cost high performance superconducting material for magnet applications. Larbalestier *et al* (2001) showed that there are no weak links in the supercurrent flow as in the high T_C oxides and that it is more similar to metallic superconductors than to the oxide high T_C materials. Its $H_{C2}(O)$ is reasonable (about 18 T) and a lot of effort has gone into making films and wires with high J_C . The J_C values achieved in thin films at 4.2 K are 10⁶ A/cm² at 1 T and 10⁵ A/cm² at 10 T (Eom *et al* 2001). MgB₂ wires with iron cladding are being tested by various groups, and J_C values reported at 4.2 K are above 10^5 A/cm² at 1 tesla; but degrade rapidly with increasing field (10^4 A/cm² at 2 tesla). These are, however, very early days and many groups are active; Wang *et al* (2001) have reported J_C values at 20 K of 10^5 A/cm² at 1.5 T and 10^4 A/cm² at about 3 T, while at 10 K they report 10^4 A/cm² at 4 T. We note that these are all inferred from M–H data. Soltanian *et al* (2001) have reported transport (pulsed) measurement of 10^4 A/cm² at 1 T and 30 K. This is clearly a promising material.

We have seen above that the only way of reducing the flux-flow resistance and enhancing J_C is to prevent the motion of vortices by pinning them with point defects or with precipitates of non-superconducting phases, a technique that is similar to the incorporation of tin impurities in copper to produce bronze (Nelson 1997). Further, the bronze age gave way to the iron age when iron was made stronger by supplementing impurities with work-hardening (Nelson 1997). He hopes that the corresponding 'bronze-age to iron-age transition' in the electrical properties of superconductors could be speeded up by ideas from basic research and fundamental science. In-depth research into the behaviour of vortices has been prompted by the peak effect in $J_C(B)$.

4. The peak effect in J_C

As noted above, those samples of YBCO that show a peak effect have a reasonable J_C in moderate fields. The peak effect holds the promise of making YBCO useful for applications at 77 K as mentioned in § 1 that the peak effect has been seen in many superconductors, and its cause has been the subject of continuing research. We discuss recent attempts to understand the peak effect, with the hope that we may then be able to use it to our advantage.

The fundamental measure of vortex pinning is the pinning force per unit volume F_P , and this dictates J_C . If F_P were roughly independent of field, then J_C would decrease with increasing field. This is what is usually observed in most material, though the decrease can be roughly as $1/(H + H_0)$ as in most conventional superconductors (Kim & Stephen 1969), or be rapid and almost exponential (as first noted in high T_C superconductors by Chaddah *et al* 1989). The peak effect is thus unexpected and its origin remains a subject of research. We wish to assert that in the present author's view the peak effect may have many causes – the concept of field-induced pinning centres has been discussed above as an explanation for the high T_C superconductors. A second possible origin, which has wider applicability, is the softening of the vortex lattice at high fields. However, different detailed pictures have been presented. Kramer (1973) proposed that the peak in F_P corresponds to a changeover in

the way flux-motion starts. At low fields the vortex motion arises due to depinning, whereas above the peak effect there is a synchronous shear of the vortex-lattice about strong pinning centres. Wordenweber *et al* (1986) argued that the peak in F_P occurs because of flux-line dislocations, and that there is a structural transition from a mainly elastically distorted to a plastically distorted lattice. These ideas are in addition to a non-local theory (Matsushita & Ekin 1994) for the elasticity of the flux-lattice, and associated lattice softening for the peak effect. Preferences for various ideas on lattice softening have been presented based on when and in what materials the peak effect is observed (Matsushita & Ekin 1994), but we shall not follow that discussion here. Finally, a third possibility is that the peak in F_P can arise if the vortex lattice in a paramagnetic superconductor transforms to the FFLO state (Tachiki *et al* 1996).

We concentrate on "history effects" in measurements of F_P (or J_C) in the region of the peak effect. These measurements have intrigued researchers for many years regarding both its possible origin and the fact that J_C (and the useful operating volume) would depend on how the sample is cooled to its operating temperature. Steingart et al (1973) first reported history effects in the pinning force when they measured J_C in a single crystal of niobium. At a given (H,T) point near the peak region they measured J_C while (a) increasing field from zero at fixed T (we shall refer to this as $J_C(ZFC)$), (b) decreasing field from above H_{C2} to H (denoted $J_C(H-)$) and (c) lowering temperature from above T_C , or field-cooling in fixed H (denoted $J_C(FC)$). They found that $J_C(FC) > J_C(H) > J_C(ZFC)$. Steingart *et al* (1973) gave the following plausible explanation for the corresponding inequality in F_P obtained following these three histories. They said that in view of the small shear modulus of the vortex lattice near H_{C2} , the vortex lattice in the field-cooled case forms in a granular structure maximising the coverage of pinning sites. This coverage persists on cooling till H, whereas in the ZFC case the vortex forms a near-perfect lattice at zero field, ignoring many of the pinning sites. This near-perfect lattice persists till H in the ZFC case, and the pinning is less efficient than in the FC case. Steingart et al (1973) also observed that the history effects disappeared above the peak effect, and explained this within Kramer's model (1973) since F_P here is dictated by the shear constant C_{66} and would be insensitive to the number of effective pinning sites. Similar history effects in the transport J_C near the peak effect region were reported in amorphous films of Nb₃Ge and Mo₃Si (Wordenweber et al 1986). It is argued (Wordenweber *et al* 1986) that F_P shows a peak because of dislocations created in the flux lattice, and involves a structural transition. The history dependence in J_C (or F_P) is ascribed to persistence, in the field decreasing case, of metastable states with a large number of dislocations.

 J_C is also inferred from M–H curves using Bean's CSM, and the existence of history effect near (below) the peak effect have been reported (Roy & Chaddah 1997) by the observation of anomalous, non-overlapping, minor hysteresis loops (MHL). In figure 3 we show the pinning force as a function of field ($h = H/H_{C2}$ is in reduced units) for CeRu₂ samples (Roy *et al* 1998), as obtained from M–H curves using Bean's critical state model. Here again the pinning force (and thus J_C) is higher in the FC case than in the ZFC case. Such observations were first reported in CeRu₂, but have since been reported in the peak effect of other superconductors including YBCO (Kokkaliaris *et al* 1999; Radzyner *et al* 2000; Zhukov *et al* 2001). The inequalities in J_C (or F_P) observed under the different histories are consistent with those observed in Nb by Steingart *et al* (1973). This flurry of activity in MHL measurements appear to have evolved a consensus (Roy & Chaddah 1997; Ravikumar *et al* 1999–2001; Roy *et al* 2000) that the history effects are due to supercooling (and superheating) across a first order phase transition in vortex matter and we shall briefly present the arguments below. It has



Figure 3. Measured pinning force vs. field (in reduced units, $h = H/H_{C2}$) for two samples of CeRu₂. The squares correspond to measurements in the field-cooled (FC) state, while the triangles correspond to the zero-field-cooled (ZFC) state. In the region below the peak effect, the pinning is stronger in the FC state.

also been noted now that history effects are not observed with every peak effect. Specific reports are on MoRe alloys (Chaddah *et al* 2001), and history effects in YBCO were shown (Kokkaliaris *et al* 2000) to vanish quite readily with change in oxygen stoichiometry and in twinned samples. Since peak effect can be originating due to various possible causes, we believe that history effects are seen only when the peak effect is associated with an underlying first order transition (Chaddah *et al* 2001).

We briefly outline the arguments associating the history effects with a FOT. Roy & Chaddah (1997) found in the PE region of CeRu₂ that MHLs initiated from the field increasing M–H curve saturated below the field decreasing envelope curve. They also found that MHLs initiated from the field-decreasing envelope curve did not close on itself. They argued that the field-decreasing state was a supercooled high J_C state and its "shattering" explained why the MHL did not close on itself. The field increasing case at H corresponded to vortex matter in phase A, and the field decreasing case to vortex matter in the supercooled phase B, with the first order transition allowing supercooling. Similar behaviour of non-merging MHLs was reported at the peak effect in NBSe₂ (Ravikumar *et al* 1999–2001) and YBCO (Kokkaliaris *et al* 1999; Radzyner *et al* 2000; Zhukov *et al* 2001; Chaddah *et al* 2001) and ascribed to a first order transition (Ravikumar *et al* 1999–2001; Zhukov *et al* 2001). Roy & Chaddah (1997) also showed a jump in the equilibrium magnetization at the onset of the PE in CeRu₂, with a sign consistent with the Clausius–Clapeyron relation, reinforcing their explanation in terms of a first order



Figure 4. Schematic of the history-dependence in J_C vs. field J_C is higher in the FC state. If the measuring current used is J_0 as indicated, then a finite resistance is seen in the ZFC state at intermediate fields, whereas no resistance is seen in the FC state (Chaudhary *et al* 2001).

transition. A similar step has been reported at the outset of PE in NbSe₂ (Ravikumar *et al* 1999–2001), again reinforcing an explanation in terms of a first order transition. The jump in equilibrium magnetisation in CeRu₂ has now been confirmed in single crystal samples (Tulapurkar *et al* 2001). Further, Chaddah & Roy (2000) have argued that supercooling can be caused by both lowering H and by lowering T, and that more persisting supercooling would be seen in the latter case for a first order transition. This has been confirmed in single crystal CeRu₂ (Roy *et al* 2000).

Further, various anomalous features seen in the MHLs have been explained semi quantitatively by calculations assuming a first order transition (Chaddah 2000; Chaddah *et al* 2001). Before concluding, we discuss an explicit example where a sample of V₃Si would offer finite resistance, or zero resistance, at the same (H, T) depending on the history in which it is cooled. Figure 4 shows a schematic of the peak in $J_C(H)$ as observed in the field-increasing ZFC case. The history effect implies that J_C observed in the FC case is larger in the field region below the peak effect. If we pass a current of density J_0 as indicated in figure 4, no resistance will be observed in the FC case, but a finite resistance will be observed in a narrow field region in the ZFC case. This is exactly what was seen V₃Si by Chaudhary *et al* (2001). They also showed that the FC state is supercooled and metastable because when a magnetic field pulse of 5 m tesla was given (in an applied field of 2.4 tesla) the supercooled FC state was shattered and the sample developed a resistance.

It thus appears that history effects near the peak effect indicate that the onset of the peak effect is associated with a first order phase transition in vortex matter. If we understand what caused the phase transition to a high J_C state, we may be able to control this transition and make it occur at a practically desired field value!

5. Conclusions

We have discussed the measurement and importance of J_C in superconducting materials. Many new superconducting materials, with attractively high values of T_C and H_{C2} have been discovered during the last fifteen years, but commercial applications are still based on low T_C intermetallics discovered much earlier. We have outlined the current status of research in two promising new superconductors. We have discussed recent research on the peak effect in $J_C(B)$ in the belief that a better understanding of vortex-matter may, in Nelson's (1997) words, allow us to "compress the time required to improve the electrical properties of superconductors".

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