# **Temperature and magnetic-field dependence of the superconducting order parameter in Zn studied by point-contact spectroscopy**

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The differential resistance  $dV/dI$  vs voltage V of Zn-Ag point contacts (PC's) is measured in the superconducting state to evaluate the temperature and magnetic-field dependence of the superconducting order parameter  $\Delta$  and  $\Gamma = \hbar / \tau$  in Zn using the modified Blonder-Tinkham-Klapwijk theory including a finite quasiparticle lifetime  $\tau$ . The temperature dependence of  $\Delta(T)$  of all PC's follows the BCS theory while for the magnetic field dependence of  $\Delta(B)$  there are marked differences among different PC's. In one case the  $\Delta(B)$  dependence shows a first-order transition at  $B_c^{PC} < B_c^{bulk}$  for other PC's  $\Delta(B)$  decreases continuously in a magnetic field and vanishes at  $B_c^{PC} \gg B_c^{\text{bulk}}$ . The parameter  $\Gamma$  which describes the broadening of the quasiparticle density of states  $(DOS)$  in the superconductor is almost independent of  $B$  in the first case, in contrast to the increase in a magnetic field in the second case. Moreover, in zero field  $\Gamma$  increases with PC resistance. Possible reasons for the DOS broadening in zero field and with increasing magnetic field or increasing PC resistance are discussed. [S0163-1829(96)02545-3]

### **I. INTRODUCTION**

The investigation of the conductivity of superconductor– normal-metal (SN) point contacts (PC's) has become a powerful tool for the study of the superconductors  $SC's$ ). The phenomenological Blonder-Tinkham-Klapwijk (BTK) model<sup>1</sup> taking into account Andreev reflection  $(AR)$  at the SN boundary provides a good description of measurements on conventional superconductors.2 The theory can explain the variation of the PC characteristics, i.e., *dV*/*dI* vs voltage *V* curves, from a clean metallic contact to a perfect tunnel junction with only one parameter, i.e., the barrier strength *Z* at the SN interface. The superconducting order parameter  $\Delta$  in the SC can be estimated and its temperature dependence is obtained from a fit according to the BTK theory.

The study of SN PC's in a magnetic field has received considerably less attention. Of course, before this method is employed for exotic SC's such as high- $T_c$  materials, organic SC's, or heavy-fermion compounds, one should check how it works with classical SC. To our knowledge there is up to now no theory of AR in a magnetic field. Likewise, detailed measurements for conventional SC's are lacking, except for  $experiments<sup>3</sup>$  where an increase of the critical magnetic field for Ta PC's was observed. Our recent investigation on Sn, In, and Nb (Ref. 4) showed that in some cases the measured curves do not correspond to the BTK model, especially for Nb. Moreover, the simple BTK theory fails to explain the dependence of the *dV*/*dI* characteristic on the magnetic field.<sup>4</sup> The main difference is the broadening of the experimental curves and the smaller amplitude of the AR feature compared to the theoretical expectation. A modified BTK theory, originally used to evaluate PC measurements in high- $T_c$  materials<sup>5,6</sup> can describe the  $dV/dI$  curves, especially their magnetic field dependence.<sup>4</sup> This model takes into account a finite quasiparticle lifetime  $\tau = \hbar/\Gamma$  caused by inelastic processes, $\alpha$  which results in a smearing of the quasiparticle density of states (DOS) of the SC.

In the present paper we have carried out detailed measurements of PC's with the classical SC Zn and used the modified BTK model to obtain the temperature and magnetic-field dependence of the superconducting order parameter  $\Delta$  along with the parameter  $\Gamma$ . Successful application of the modified BTK model to a classical SC in our opinion renders PC spectroscopy very useful for investigations of more complicated SC's. Moreover, these measurements are interesting to compare with our results on  $Zn-UPt_3$  contacts<sup>8</sup> which were carried out in order to look for a possible influence on AR of the different conditions at the SN boundary with heavyfermion metals.

#### **II. EXPERIMENT**

The samples were mounted inside the mixing chamber of a  ${}^{3}$ He/ ${}^{4}$ He dilution refrigerator. Mechanical feedthroughs and a differential screw mechanism allowed to establish PC's by letting a polycrystalline Zn wire (diameter 0.2 mm) approach and contact an Ag surface directly in the mixing chamber. The lowest measuring temperature was 30–50 mK. The applied magnetic field produced by a superconducting magnet was normal to the main current flow through the PC. The differential resistance *dV*/*dI* vs *V* was measured by the





usual lock-in technique in the temperature range between 0.03 and 1 K. Of the 24 different PC's investigated, 10 were studied in detail to find the temperature and magnetic-field dependence of *dV*/*dI* vs *V*. All PC's were established on the same Zn wire by mechanically scanning it over the Ag surface before establishing the contact.

# **III. RESULTS**

Figures 1 and 2 display the temperature and magneticfield dependence of the *dV*/*dI* curves vs *V* for two Zn-Ag PC's with almost the same resistance. The curves show a distinct double-minimum structure which changes smoothly with increasing temperature. In a magnetic field *B* we observe two different types of behavior. Either the *dV*/*dI* curves evolve smoothly towards the normal state with increasing  $B$  [Fig. 2(b)], or the double-minimum structure abruptly disappears with increasing *B* between  $B_c^{PC}$ =4.06-4.15 mT [Fig. 1(b)], which is slightly smaller than the bulk critical field  $B_c^{\text{bulk}} \sim 5.5$  mT. This doubleminimum structure is a hallmark of AR at the SN interface of a metallic PC with a small barrier, i.e.,  $0 < Z < 1$ .<sup>1</sup> Of the 10 contacts studied in detail, four (nos. 2.4, 6, and 10, see Table I) showed the discontinuous behavior with  $B_c^{\text{PC}}$  between 4.1



FIG. 1.  $dV/dI$  curves vs *V* (dots) for a Zn-Ag contact (no. 2) with  $R_N = 0.5 \Omega$  and  $T_c = 0.82 \text{ K}$ (a) for different temperatures  $T$  and (b) in different magnetic fields *B*. Fit parameters for the theoretical curve (dashed line) at  $T=0.06$  K are  $\Delta$ =110  $\mu$ eV,  $\Gamma$ =6  $\mu$ eV, and *Z*=0.5. The fit curves are almost indistinguishable from the data also for higher temperatures and are therefore not shown.

and 4.6 mT, and the other six went normal at fields of 8–26 mT with a continuous suppression of the AR feature. Similar differences in the field dependence of PC spectra were already observed in In-Ag and Sn-Ag contacts without, however, a systematic investigation.<sup>4</sup>

For the description of the measured curves and their temperature and magnetic-field dependences, the modified BTK model was used. This model includes finite-lifetime effects described by a parameter  $\Gamma$  resulting in a broadening of the quasiparticle DOS in the SC:6

$$
N(E,\Gamma) = \text{Re}\left\{\frac{\varepsilon + i\Gamma}{\left[(\varepsilon + i\Gamma)^2 - \Delta^2\right]^{1/2}}\right\}.
$$

The data can be described very well with this model (cf. dashed lines in Figs. 1 and 2 for curves at  $0.06 \text{ K}$ ). A complete fit, i.e., without adjusting the amplitude of the measured and theoretical curves, was only possible by admitting  $\Gamma$  $\neq$ 0, especially for curves in a magnetic field. The barrier parameter  $Z$  was kept fixed, independent of  $T$  and  $B$ .  $\Gamma$ , too, was found to be independent of *T* for all PC's under investigation. The temperature dependence of  $\Delta(T)$  for Zn obtained by the fitting procedure is shown in Fig. 3.  $\Delta(T)$  for a PC with  $2\Delta(0)/kT_c \approx 3.5$  is in perfect accord with the BCS

FIG. 2.  $dV/dI$  curves vs  $V$  (dots) for a Zn-Ag contact (no. 1) with  $R_N = 0.47$   $\Omega$  and  $T_c = 0.83$  K (a) for different temperatures  $T$  and (b) in different magnetic fields *B*. Fit parameters for the theoretical curve (dashed line) at  $T=0.06$  K are  $\Delta = 124$   $\mu$ eV,  $\Gamma = 6.5$   $\mu$ eV, and  $Z = 0.51$ .

No.	$R_N(\Omega)$	$T_c$ (mK)	$B_{c2}^{\rm PC}$ (mT)	$\Delta(0)$ ( $\mu$ eV)	Ζ	$\Gamma$ ( $\mu$ eV)	$2\Delta(0)/kT_c$
$\mathbf{1}$	0.47	830	8	124	0.51	6.5	3.47
2	0.5	820	4.15	112	0.5	6	3.17
3	0.6	815	26	130	0.46	9	3.7
$\overline{4}$	0.87	820	4.42	105	0.47	10	3.0
5	0.98	835	15	102	0.47	7	2.8
6	1.19	840	4.62	125	0.5	10	3.45
7	2.03	850	26	135	0.42	28	3.7
8	2.19	815	11	135	0.42	27	3.84
9	2.5	815	25	105	0.4	23	3.0
10	2.84	825	4.4	120	0.45	27	3.37
Average		$826 \pm 12$		$119 \pm 12$			$3.35 \pm 0.35$
Ref. 12		875	5.3	120			3.2

TABLE I. Parameters of the measured Zn-Ag PC's. See text for symbols.

theory.  $\Delta(T)$  for a PC with  $2\Delta(0)/kT_c \approx 3$  is somewhat below the BCS curve whereas for a PC with  $2\Delta(0)/kT_c$   $\simeq$ 3.8,  $\Delta(T)$  is above.

The variation of  $\Delta(B)$  and  $\Gamma(B)$  in a magnetic field is shown in Fig. 4. Again, two distinct types of behavior are observed. In one case, corresponding to the contact of Fig. 1(b),  $\Delta(B)$  remains almost constant with increasing magnetic field until a sudden drop at  $B_c^{PC} < B_c^{bulk}$  indicates a firstorder transition to the normal state. For other PC's such as that of Fig. 2(b)  $\Delta(B)$  decreases continuously with increasing  $B$  [Fig. 4(b)] showing for some PC's a dependence characteristic of a thin superconducting film in a parallel magnetic field,  $\Delta(B) \propto [1 - (B/B_c)^2]^{1/2}$  (Ref. 9).

The parameter  $\Gamma$  increases only slightly with *B* in the first case, while in the second case  $\Gamma$  increases considerably with



FIG. 3.  $\Delta(T)$  for different PC's with  $2\Delta(0)/kT_c = 3.0 \pm 0.1$ (top),  $3.45 \pm 0.05$  (middle), and  $3.75 \pm 0.05$  (bottom). Different symbols indicate different PC's. Data are normalized by  $\Delta_0 = \Delta(0)$ .

magnetic field. We have found that  $\Gamma(B=0)$  increases also with the PC resistance (Fig. 5, Table I). The zero-field  $\Gamma$  and its increase with PC resistance are a few times lower as was found for  $Zn$ -UPt<sub>3</sub> PC's.<sup>8</sup>

#### **IV. DISCUSSION**

 $\Delta(T)$  for Zn perfectly follows the BCS theory for PC's with  $2\Delta(0)/kT_c$  close to the theoretical BCS value 3.5. PC's with  $2\Delta(0)/kT_c \approx 3.8$  (above this value) show a  $\Delta(T)$  dependence slightly above the BCS curve as found in the case of strong-coupling SC such as Pb or some alloys.<sup>11</sup> Consequently, PC's with  $2\Delta(0)/kT_c \approx 3$  (below the theoretical value) show a  $\Delta(T)$  dependence below the BCS curve. The agreement of the  $\Delta(T)$  dependence with the BCS theory provides evidence that the modified BTK model has a solid physical foundation and can be used for the determination of, e.g., the magnetic-field dependence of  $\Delta$ .

In the majority of cases the value of the barrier strength *Z* was found to be between 0.4 and 0.5 (Table I). Normal reflection of the quasiparticles at the interface caused by mismatch of the Fermi velocities  $v_F$  yields  $Z \approx 0.13$  using  $Z = (1 - r)/2r^{1/2}$  (Ref. 1), where  $r = v_{F1}/v_{F2}$  and  $v_F^{\text{Ag}} = 1.39 \times 10^6$  m/s and  $v_F^{\text{Zn}} = 1.82 \times 10^6$  m/s.<sup>12</sup> Thus some natural barrier (e.g., oxide) is always present in the PC. It is



FIG. 4.  $\Delta(B)$  (triangles) and  $\Gamma(B)$  (rectangles) at  $T \approx 0.06$  K for  $PC's$  (a) from Fig. 1 and (b) from Fig. 2. Solid and dash-dotted lines in (b) are  $\Delta \propto [1 - (B/B_c)^2]^{1/2}$  [as for thin films (Ref. 9)] and  $\Delta$  $\propto (1-B/B_c)^{1/2}$  [as for type-II SC's (Ref. 10)]. Dashed lines are guides to the eye.



FIG. 5. Dependence of the parameter  $\Gamma$  on the PC resistance  $R_N$  for Zn-Ag and Zn-UPt<sub>3</sub> (Ref. 8) contacts. Solid lines are guides to the eye.

interesting to note that the same *Z* value is often found both for high- $T_c$  materials<sup>6</sup> and for heavy-fermion superconductors.<sup>8</sup> This indicates that the difference in the Fermi velocities plays a negligible role in all these cases and some physical barrier is at the origin of *Z*.

The two distinct types of behavior of the  $\Delta(B)$  and  $\Gamma(B)$  dependence in a magnetic field observed for Zn (Fig. 4) are connected, according to our earlier suggestion, $4$  to different structures of the metal in the PC. For a type-I SC the transition into the normal state is of first order with a discontinuous drop to zero of the energy gap at  $B_c^{\text{bulk}}$ . In our case the critical field in the PC is smaller than  $B_c^{\text{bulk}}$  presumably because of the demagnetization effect arising from the geometry of the PC.

A second-order transition occurs for a type-I SC only if it is a thin film with thickness  $d < 3\lambda_L$ , where  $\lambda_L$  is the London penetration depth.<sup>9</sup> Indeed, the  $\Delta(B)$  dependence in Fig. 4(b) looks like the  $\Delta(B)$  behavior of a thin superconducting film. In case of disorder in the PC resulting in a short mean free path  $l \ll \xi_o$  the SC might be of type II, i.e.,  $\xi \approx (\xi_0 l)^{1/2} < \sqrt{2}\lambda \approx \sqrt{2}\lambda_L(\xi_0 / l)^{1/2}$ . This yields an increase of the critical field  $B_{c2} \simeq \Phi_0 / \xi^2$  and a second-order transition to the normal state. The calculated values of  $\xi$  and  $\lambda$  (Table II) show that indeed  $\xi < \sqrt{2\lambda}$  for all PC's with second-order transition.

It should be noted that we observed the drastically differ-

TABLE II. Calculated characteristic lengths for the PC's with second-order transition assuming  $\xi_0 \approx 2000$  nm and  $\lambda_L \approx 30$  nm for Zn (Ref. 15), using  $l \approx \Phi_0/2\pi \xi_0 B_{c2}$ ,  $\xi \approx (\xi_0 l)^{1/2}$ , and  $\lambda \approx \lambda_L (\xi_0 / l)^{1/2}$ . *d* is the PC diameter calculated according to the equation  $R_N \approx \rho/d + 16\rho l/3\pi d^2$ , where  $\rho l \approx 0.5 \times 10^{-11} \Omega$  cm<sup>2</sup> from Ref. 13.

No.	$R_N(\Omega)$	$B_{c2}^{PC}$ (mT) $l$ (nm) $\xi$ (nm) $\lambda$ (nm) $d$ (nm)				
1	0.47	8	21	203	293	75
3	0.6	26	6	113	548	148
5	0.98	15	11	148	404	61
7	2.03	26	6	113	548	50
8	2.19	11	15	173	346	29
9	2.5	25	7	115	507	38
Bulk		5.3	$\geq 100$	2000	30	

ent  $\Delta(B)$  dependences for PC's with the same resistance or almost of equal size. This means that a size effect  $(e.g.,$  as caused by an increase of the parallel critical field for thin films) plays a negligible role in our case, leading us to suppose that the different types of structures of the metal in the constriction cause the different behavior. Moreover, we have observed the first-order transition for PC's with resistance up to 10  $\Omega$  corresponding to a linear dimension of about 10 nm according to Eq.  $(10)$  in Ref. 13, or a few times *less* than  $\lambda_L$ <sup> $\approx$ </sup> 30 nm. This is once more in contrast with the hypothesis of a thin film of type-I SC where the first-order transition is observed only for  $d \ge 3\lambda_L$ .<sup>9</sup>

Interestingly, the first-order transition was observed only for PC's with  $2\Delta(0)/kT_c \leq 3.5$ , whereas PC's with a secondorder transition have smaller as well as larger values of the reduced gap that 3.5. The latter observation suggests different reasons for the reduction of the mean free path. First, the strong structural disorder in the PC may result in a softening of the phonon spectrum of the metals leading to an enhanced electron-phonon coupling parameter  $\lambda$  corresponding to a strong-coupling SC. Second, impurity contamination of the PC does not necessarily change the phonon spectrum and electron-phonon interaction, and hence may result only in a reduction of the mean free path without altering the reduced gap.

Turning to a discussion of  $\Gamma$ , we first note that  $\Gamma$  for  $B=0$  is often attributed to inelastic scattering, i.e.,  $\Gamma = \hbar / \tau_{\text{in}}$ , with the inelastic scattering rate  $\tau_{\text{in}}^{-1}$ .<sup>7</sup> However, in our case it is difficult to associate  $\Gamma$  with "usual" inelastic scattering processes such as electron-phonon and electronelectron interactions because these processes are negligible at the low temperatures of our experiment. Figure 5 shows that  $\Gamma$  increases roughly linearly with the PC resistance  $R_N$ while at the same time  $\Delta(0)$  (not shown) shows a large amount of scatter varying between 105 and 135  $\mu$ eV (Table I), but within this scatter is independent of  $R_N$ . There is no correlation between  $\Delta(0)$  and  $\Gamma$  which might be expected if  $\Gamma(0)$  is related direct to pair breaking.

It should be emphasized that Zn is an anisotropic metal because the *c*/*a* ratio deviates from the ideal value 1.63 for a hexagonal close-packed structure.<sup>12</sup> This leads to a strong anisotropy of the electron-phonon interaction spectra<sup>13</sup> which may result in a gap anisotropy. In this case some gap distribution in our polycrystalline PC is possible, which can be described effectively by  $\Gamma$ . However, in this way it is difficult to explain the increase of the  $\Gamma$  with PC resistance while  $\Delta(0)$  is independent of  $R_N$ . Because all our PC's could be modeled with a lifetime-broadened DOS, a gap distribution would imply the same type of distribution function for all PC's, which is unlikely. Hence the main origin of a finite  $\Gamma$  at  $B = 0$  is unclear. Furthermore, there is no correlation between the zero-field  $\Gamma$  and the type of superconductor - normal-metal transition in a magnetic field.

For the PC's with type-II SC's we observed that  $\Gamma$  increases significantly in a magnetic field. Indeed, for pairbreaking processes which do result in a smearing of the DOS at the gap edges it is known that the pair-breaking parameter  $\alpha$  increases in a magnetic field and  $\alpha \sim B$  for a type-II SC.<sup>10</sup> Moreover Strässler and Wyder showed<sup>14</sup> that smearing of the DOS of a thin film in a parallel magnetic field depends on the mean free path *l*. For large *l* this DOS is similar to the DOS in the modified BTK theory described by  $\Gamma$ . Hence pair-breaking processes might qualitatively explain the increase of  $\Gamma$  with *B*.

The zero-field  $\Gamma$  and its increase with the PC resistance are a few times higher for  $Zn$ -UPt<sub>3</sub> PC's (Ref. 8) than for Zn-Ag PC's (Fig. 5). Furthermore,  $\Gamma$  is considerably higher for PC's with Nb (Ref. 4) and especially for high- $T_c$ materials<sup>6</sup> suggesting that the conditions of the SN interface strongly influence the pair-breaking processes. The short coherence length in the latter materials might also play a role.

## **V. SUMMARY**

We have observed unmistakably in SN point contacts the first-order transition of a type-I SC to the normal state in a magnetic field. The second-order transition occurring in some PC's is attributed to the shortening of the SC coherence length via a reduction of the mean free path in the PC. This leads to type-II superconductivity with an increased critical magnetic field.

The modified BTK model can be used to reliably determine from the *dV*/*dI* curves of PC's the temperature and magnetic-field dependence of the order parameter  $\Delta$  and of the lifetime parameter  $\Gamma$  describing the quasiparticle DOS smearing of the SC.

The  $\Delta(T)$  dependence corresponds well to the BCS

theory.  $\Delta(B)$  shows a discontinuous drop at the critical field for ''clean'' PC's indicating a first-order transition while for "dirty" PC's  $\Delta(B)$  decreases gradually in a magnetic field. Correspondingly, the quasiparticle DOS of the SC remains almost unaffected in the first case whereas for PC's with a second-order transition the DOS is smeared in a magnetic field. The latter is expected for a type-II SC because of the increase of the pair-breaking parameter with *B*. 9

The finite zero-field value of  $\Gamma$  depends on the PC resistance and also on the material for both SC and normal metal. The fact that  $\Gamma$  is independent of temperature appears to exclude conventional inelastic scattering processes. One can only speculate about a possible origin such as impurityinterface scattering, gap anisotropy-distribution or a strongly nonequilibrium state in the PC.

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