Analysis of point-contact Andreev reflection spectra in spin polarization measurements

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We present a systematic analysis of point-contact Andreev reflection (PCAR) spectra for ferromagnetic materials, using both modeling and experimental data. We emphasize the importance of consistent data analysis to avoid possible misinterpretation of the data. We consider the relationship between ballistic and diffusive transport, the effect of different transport regimes on spin polarization measurements, and the importance of unambiguous identification of the type of transport regime. We find that in a realistic parameter range, the analysis of PCAR spectra of purely diffusive character by a ballistic model yield approximately the same (within ~3%) values of the spin polarization and the barrier strength Z larger by ~0.5–0.6. We also consider the dependence of polarization values on Z, and have shown by simple modeling that letting the superconducting gap vary as an adjustable parameter can result in a spurious dependence of the spin-polarization P_c on Z. At the same time we analyzed the effects of finite Z on the apparent value of P_c measured by the PCAR technique, using a large number of examples from both our own measurements and from the literature. We conclude that there is a system-dependent variation in $P_c(Z)$, presumably due to spin-flip scattering at the interface. However, the exact type of this dependence is hard to determine with any statistical certainty.

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Spin-polarized ferromagnetic materials are set to play a key role in the next-generation electronic devices, based on the electron's spin rather than charge.[1] The performance of many of these *spintronics* devices improves dramatically as the spin polarization P of the ferromagnetic material increases.[1] Particular attention has focused on the so-called "half-metals," in which the electrons responsible for the metallic transport all have the same spin (either spin up or spin down), while the electrons with the opposite spin are insulating. Half-metals have the maximum attainable value of spin polarization ($P=1.0$). Most of the experimental studies to determine P have been carried out by the spin-dependent tunneling technique, pioneered by Tedrow and Meservey.[2] This method requires the material of interest to be fabricated as part of a ferromagnet/superconductor tunnel junction, in which the superconducting density of states is then Zeeman-split by the application of a magnetic field of several tesla. The other conventional technique is spin-resolved photoemission, which measures the spin of the electrons emitted from a region close to the surface of the ferromagnet of the order of 5–20 A, and thus is quite surface-sensitive.[3]

The point contact Andreev reflection (PCAR)[4,5] technique serves to expedite and widen the search for many new materials that are too difficult to incorporate into tunnel junctions. PCAR, which is deceptively easy to put into practice, is a technique in which the conductance ($G \equiv dI/dV$) is measured for an electrical point contact with little or no tunneling barrier established between a superconducting tip and a ferromagnetic counter-electrode (or *vice versa*). The presence of spin-polarized current in the ferromagnet alters the conductance of the contact in a known way, giving rise to a new technique to determine the spin polarization P_c. This method offers several apparent advantages compared to the other techniques. With no restrictions on the sample geometry, one can avoid complex fabrication steps. In addition, it has excellent energy resolution (~1.0 meV), and does not necessarily require an applied magnetic field. However, as with any new technique, care should be taken to ensure that the resulting values of spin polarization are meaningful. There are some objective difficulties, such as possible surface modification (unless all the measurements are done *in situ*), due to uncontrolled surface oxides, or other chemical reactions on the surface of both the ferromagnet and the superconductor. The effect of this surface modification on P_c is difficult to quantify; however, in many cases the fragile surface oxide layer can be penetrated as the point contact is established, thus circumventing this problem. Care should also be taken to prevent any excessive pressure from the superconducting tip to the sample, as this may potentially change the electronic properties of the material thus affecting the values of the spin polarization. It is not always possible to directly compare the PCAR results to the values of the spin polarization obtained by other methods where the surface oxidation is better controlled (Ref. 2), as the definition of the spin polarization measured in different experiments do not necessarily coincide, as we will discuss in more detail below.

Most importantly, a comprehensive analysis of the PCAR data is nontrivial, especially compared with the simple experimental conductance measurement technique, which is used to obtain the data. This analysis, which will be addressed in this work, should include independent determination of the type of the contact (ballistic versus diffusive) as well as the main point contact parameters. The most impor-

tant parameters are the superconducting gap, the transparency of the junction, and, especially in the case of thin films, the spreading resistance of the sample, which can either be estimated during the conductance measurements or measured separately using a standard four-probe technique. As we will show below variations of these parameters can strongly affect the extracted values of P_c and their evaluation should be an inherent part of the technique, if applied correctly.

PCAR spectra to date have been typically analyzed using the Blonder-Tinkham-Klapwijk (BTK) model,[6] modified to include a spin polarization of the metal.[4,7] It is a weak coupling theory describing all interface effects by a single dimensionless parameter Z, which does not necessarily correspond to any physical parameter characterizing the interface barrier. Recently, the applicability of the BTK and the modified BTK formulas to the spin polarization measurements has been questioned.[8] Undoubtedly the BTK theory does not properly take into account some of the delicate surface phenomena, such as the presence of surface states and the effects of lattice relaxation. The theory also makes assumptions the validity of which are difficult to evaluate, such as the δ-functional form for the barrier, the step-function shape of the voltage drop across the barrier, and lateral momentum conservation. In addition, the modified BTK model also assumes spin-independent barriers.[4,7] Xia *et al.* (Ref. 8) performed advanced local-density approximation calculations for Andreev transport across realistic interfaces. These calculations took care of some, but not all of the issues listed above. However, they were not able to satisfactorily describe experimental curves[5] of ferromagnetic systems, while the modified BTK formulas, treating Z as an adjustable parameter, provide an excellent description of the same curves. Although the formalism of Ref. 6 is based on a derivation where a δ-shaped barrier is assumed, Z actually incorporates more physics than just the strength of the δ-function, and, therefore, the formalism works much better than could have been expected.[9] That Z is not the real barrier strength in actual measurements is emphasized by the fact that sometimes the BTK model fits experimental curves surprisingly well with $Z=0$, although formally, due to the Fermi velocity mismatch between the metal and the superconductor, there always exists some minimal nonzero Z. Probably the most illustrative case is that of the colossal magnetoresistance material $La_{1-x}Sr_xMnO_3$ (LSMO),[10] where because of the large disparity between the Fermi velocities of the majority and minority spin electrons, at least in one spin channel the Fermi velocity mismatch should be very large.[11]

Another important question which was raised in connection with the BTK formalism is the difference between the ballistic and diffusive transport, defined by the ratio of the mean free path l of the electrons and the contact diameter d. In general, there are *three* possible types of transport in a PCAR experiment: ballistic ($l \gg d$), diffusive ($l \ll d$), and intermediate ($l \sim d$). One way of estimating the mean-free path is from the Drude formula, using a measured value for conductivity σ ($\sigma = ne^2 l/mv_F$), where (n/m) and v_F can be calculated from the band structure. The diameter of the contact d can then be calculated employing the equation for the junction (contact) resistance[12]

$$R_N = R_0(1+Z^2) \approx (4\rho l/3\pi d^2 + \rho/2d)(1+Z^2), \qquad (1)$$

where the first term in the expression for R_o is the so-called Sharvin resistance[13] for ballistic contacts, while the second is the Maxwell resistance[14] for diffusive transport. To determine d, R_N must be measured and Z obtained by analyzing the conductance curves. Alternatively, one can estimate the size of the contact d and the mean free path independently (and potentially more accurately) by experimentally measuring the contact resistance in a broad temperature range, which, however, requires high thermal stability of the contacts.[15] In many cases, it is hard to avoid fairly large uncertainties in making such estimates and thus it is often difficult to establish the exact transport regime for the junction conduction. Additionally, the ratio of l/d can often be close to one, thus indicating the transport regime in the intermediate region. The applicability of the ballistic theory for the data obtained in this regime, especially given the uncertainty in estimating l and d, may seem problematic.

According to Ref. 7, it is possible to extend the BTK theory onto the diffuse limit. There is an issue as to which model should be applied to a given set of data. Moreover, no theory has yet been developed for the intermediate case. It is believed that both the ballistic and diffusive formalisms will yield approximately the same value for P_c, albeit with different Z values, when analyzing the same spectra. If so, and assuming that the behavior for the intermediate case is bracketed by the ballistic and diffusive limits, then it does not really matter which regime applies to a particular junction in a particular experiment, as long as the value of Z is not of interest (one should, however, be aware that the current spin polarization of a material may be different for the ballistic and diffusive transports[19]). This fact may explain why several different groups, exercising no particular control over the transport regime for their point contacts, and using only the ballistic model for the analysis (i.e., implicitly assuming the ballistic regime), still obtained comparable results for P_c for the same materials. Earlier attempts to analyze the same experimental spectra with both diffusive and ballistic formulas seemed to indicate that the resulting polarizations are very close;[11] however, no systematic tests of this assumption have been performed.

In this paper we will present an analysis of PCAR spectra in both the ballistic and diffusive limits for several ferromagnetic oxides, mainly CrO_2 and LSMO, in order to illustrate some possible caveats in applying the modified BTK formulas to real materials. First, we will discuss the differences between the diffusive and the ballistic models. Second, we will consider the sources of possible systematic errors, when analyzing $G(V)$ curves. Specifically, we will discuss the effect of using the superconducting gap Δ as a variable parameter on the extracted value of P_c and show how, with the inclusion of the additional spreading resistance R_s of the sample at a given experimental temperature, the effects of Δ on P_c can be eliminated. Finally, we will consider the possible P_c versus Z dependence seen frequently in PCAR spectra by performing a systematic analysis of a large number of different experiments. It has been argued that the functional dependence of $P_c(Z)$ is quadratic,[10,16,17] or exponential.[18] Us-

TABLE I. The total interface current in different regimes. The following notations are used: $\beta = V/\sqrt{|V^2-\Delta^2|}$, $F(x) = \cosh^{-1}(2Z^2+x)/\sqrt{(2Z^2+x)^2-1}$.

	$eV<\Delta$	$eV>\Delta$
Ballistic nonmagnetic	$\frac{2(1+\beta^2)}{\beta^2+(1+2Z^2)^2}$	$\frac{2\beta}{1+\beta+2Z^2}$
Ballistic half-metallic	0	$\frac{4\beta}{(1+\beta)^2+4Z^2}$
Diffusive nonmagnetic	$\frac{(1+\beta^2)}{2\beta}\mathrm{Im}[F(-i\beta)-F(i\beta)]$	$\beta F(\beta)$
Diffusive half-metallic	0	$\beta F[(1+\beta)^2/2-1]$

ing statistical analysis, we will show that either exponential, quadratic, or linear dependence has no apparent advantages over one another.

As we have mentioned above, two different models, ballistic and diffusive, may be used to extract values of P_c from the data for $G(V)$. Both models separate the current at the N/S interface into spin-polarized and nonpolarized contributions, and give the expressions for $G(V)$ for the two transport regimes in terms of the superconducting order parameter Δ, the bias voltage, and the interfacial barrier strength Z. Table I[7] shows the equations for the total current at the interface. In addition, the equations that describe the conductance also contain prefactors in terms of the density of states N at the Fermi level and the Fermi velocity v of both majority and minority spins. For the ballistic case, the pre-factors are $\langle Nv\uparrow,\downarrow\rangle$, while those for the diffusive are $\langle Nv^2\uparrow,\downarrow\rangle$. In practice, for both models one also needs to include corrections for the spreading resistance of the sample R_s, the additional resistance of the sample between the junction, and one of the electrical contacts in a typical four-probe measurement scheme. The presence of R_s results in the shift of the apparent position of the coherence peak $G(V)$ from $V\approx\Delta$ to larger voltages and in the change of the observed zero bias conductance value. These effects have to be always taken into account, unless R_s is much lower than the contact (junction) resistance, which is often the case only for bulk samples or highly conductive films.[11,12] Δ and R_s can, in principle, be used as fitting parameters or can be determined experimentally. We believe that it is the latter approach that is appropriate, as was done consistently in this paper. The temperature of the contact is another one of the experimental parameters that should be measured independently. A separate issue that will be addressed below in more detail is the case of the spectral broadening, which can take place for a number of reasons, one of them being a possible local heating effect in the region of the contact.[23]

First we pose the following question: *If an experimental PCAR spectrum with zero or finite Z is in reality obtained in the diffusive regime, can one apply the ballistic, rather than diffusive model to analyze it*? Furthermore, if this is possible, how will the values of the parameters (P_c and Z) compare? To answer this question, we first generated a large number of $G(V)$ curves for a hypothetical superconductor using the diffusive model with given values of P_d and Z_d (diffusive spin-polarization and barrier strength, respectively). We then analyzed these curves using the ballistic model to obtain "ballistic" values of the spin-polarization P_b and the barrier strength Z_b. The values of T, Δ, and R_s were 0.1 or 1.5 K, 1 meV, and 1.0 Ω, respectively and these values were kept throughout. The results for this procedure are shown in Figs. 1(a) and 1(b) where we plot P_b-P_d and Z_b-Z_d versus P_d for values of $Z_d=0.0$ and 0.75. The two main points illustrated by Fig. 1 are that: (1) Although this procedure tends to overestimate P_c for small polarizations, and slightly underestimate it for $P_c\sim 0.6-0.8$, potential errors introduced by applying the ballistic formulas to the diffusive contacts are negligible, less than ±3% in absolute value, for most of the

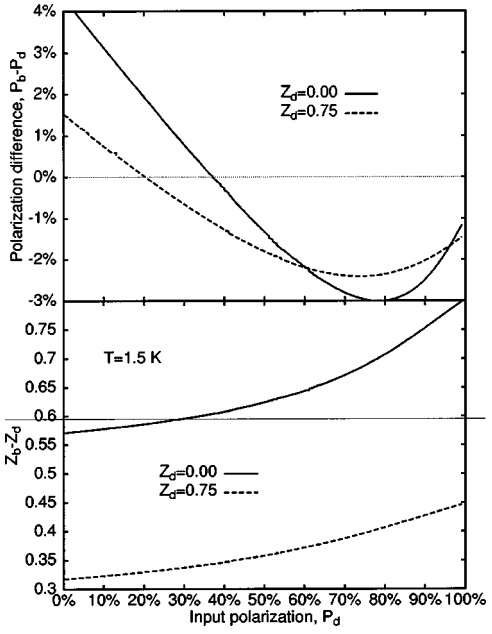

FIG. 1. Comparison between the assigned values of the spin-polarization and Z parameters using the diffusive model (P_d,Z_d) and the fitted values using the ballistic model (P_b,Z_b). The two vertical axes show the difference in (a) P_b-P_d and (b) Z_b-Z_d. The largest difference between the polarization values is 5%.

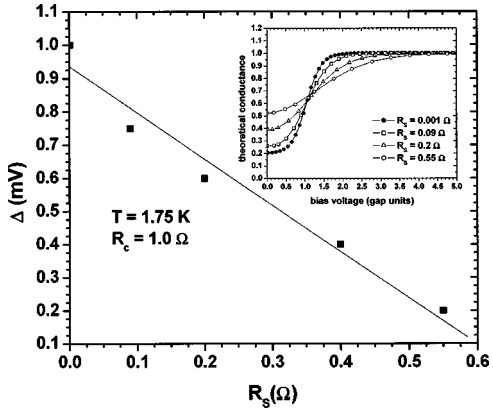

FIG. 2. Plot of Δ versus R_s from theoretically generated curves. The temperature used to generate the curves was the same as that in a typical experiment ($T=1.75$ K). The current used to generate the curves contained a contact junction resistance of $R_c \cong 1.0\ \Omega$. The inset shows some of the theoretical curves generated with values of R_s.

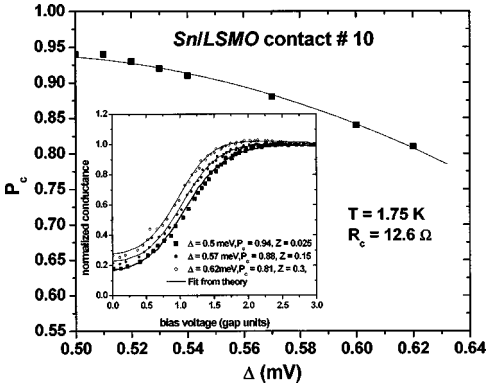

FIG. 3. P_c versus Δ for one point-contact spectrum of Sn/LSMO. The inset serves to illustrate that, in each extracted value of P_c obtained from the models, a quality fit was achieved. All fits were done in the ballistic limit. The value of the contact junction resistance for this point contact was $R_c \cong 12.6\ \Omega$, which is the contact junction resistance without subtracting out R_s. The limits for R_s in the datasets (inset) for values of Δ between 0.5–0.62 meV are approximately 11.8–9.3 Ω, respectively.

spin polarization range (2) whereas the obtained values of Z differ significantly for the two models. A comparison between the values of Z_b and Z_d for all fits showed that Z_b is always greater; for small Z_d and P_d the difference is 0.5–0.6. This is one of the illustrations of the hidden power of the BTK model: In our diffusive case, where we do have a δ-functional barrier with a *known* strength, *plus* another physical effect, not accounted for in the original BTK formalism, we see that the single parameter Z absorbs all this additional physics, producing practically the same values of the spin polarization. The main corollary of this is that when analyzing an *individual spectrum* one need not know which regime the contact was in. The uncertainty will be translated into an uncertainty in Z, but not P. Unfortunately, this property is lost, as discussed later in more detail, when a set of spectra is analyzed and an interpolation to a hypothetical $Z=0$ contact is used.

The second question we pose is the following: *What is the effect of changing the value of the superconducting gap on the extracted values of the spin polarization*? As we have mentioned above, it has become a rather common practice in PCAR studies to take a succession of G (V) curves for different point contacts and to analyze each one of them to obtain the values of P_c and Z. Thereafter one plots P_c versus Z, which is then extrapolated to $Z=0$ to obtain an "intrinsic" value of P_c for the system.[10,16–18] However, quite often the coherence peak is displaced from its theoretical position near the bulk superconducting gap. This effect can have two different causes: variation of the superconducting gap near the interface and/or the presence of the spreading resistance R_s. In fact, both Δ *and* R_s in every experiment should be uniquely determined. R_s can be measured independently, whereas Δ can be inferred from T_c using the BCS model or the experimental data for bulk superconductors. However, in many cases the analysis is done using Δ as an *adjustable parameter*, which, as we will show below, can strongly affect the values of the spin-polarization. Varying Δ is related to varying R_s, in the sense that both shift the apparent coherence peak from its BCS value, albeit in the opposite directions.

To better illustrate the relationship between Δ and R_s, we generated several theoretical curves for the *same* Δ, spin-polarization, and Z, but with different R_s in Fig. 2 (inset). This imitates an experimental situation when several contacts with different R_s are measured. However, we can now describe the same set of curves using the *same* R_s if we allow the gap to vary from curve to curve. Figure 2 shows the resulting dependence of Δ on R_s. Importantly, now the two other parameters, P and Z are also different for different curves. In other words, by analyzing experimental data collected with different R_s without taking the spreading resistance properly into account (by using incorrect values R_s or no resistance whatsoever), a wrong Δ is obtained and this error propagates into the value of the spin-polarization (see Fig. 3).

Obviously, spurious dependencies appear in such a case for all three quantities: Δ, Z, and P. This can be easily mistaken as a *dependence* of P on Z, as we show in Fig. 3. There we used a *single* point contact spectrum of Sn/LSMO with the experimentally measured $R_s=10\ \Omega$ at $T=1.75$ K with the resistivity of the LSMO film approximately 500 $\mu\Omega$ cm at 4 K. If we now assume different values for R_s, the conductance curves G (V) as a function of voltage at *the point contact* will be different (in other words we generate a whole series of curves based on a single experimental curve, see the inset in Fig. 3). We then analyzed each of the resulting curves using the standard BTK formulas and find a *different* value Δ and for P_c for each curve (the change in Δ is not surprising, since as we have shown in Fig. 2 change in R_s and Δ are related). As we can now see from the plot, 1% error in Δ corresponds to approximately 1% error in P_c. It is always desirable, therefore, to evaluate the gap and the spreading resistance separately. If this is not possible, it may be prudent to fix the value of Δ, rather then let it vary as an additional parameter. However, if the apparent position of the coherence peak in the raw spectra is shifted to significantly smaller values than the bulk gap, it may be an indica-

tion of a surface suppression of the order parameter, in which case more elaborated models are needed.

That brings us to another important point, namely, whether or not the dependence of P_c on the value of Z, often reported in the literature, is real. As one can see from Fig. 3, in this case there is a clear correlation not only between Δ and P_c, but also between the value of the Z-parameter and P_c, as Z is also monotonically changing with the gap. On the other hand, we *know* from the onset, that the actual data in Fig. 3 corresponds to just one Z (the same way it corresponds to a single value of R_s), so the "observed" Z-dependence is utterly spurious. Note that the limiting value of P_c at $Z=0$ in this case is not necessarily the "intrinsic" spin polarization, as both the gap value and R_s corresponding to this Z may be incorrect. However, we do not want to leave the reader with a conclusion that all of the observed P_c (Z) dependencies are artifacts, and, as we will show below, in a number of cases we did observe this dependence, in spite of all possible precautions in analyzing the data.

To further discuss the two models, we present Pb/CrO_2 and Sn/CrO_2 data, which are analyzed in both the ballistic and diffusive limits. The (100) CrO_2 films used in this study were made by the chemical vapor deposition method described extensively elsewhere.[20] Resistivity of these samples was found to be approximately 20 $\mu\Omega$ cm at $T=4.2$ K, somewhat higher than other reported values.[20] The measurements of these samples were done soon after the film deposition in order to avoid film degradation. The measurements with both Pb and Sn contacts were performed in a liquid He bath at temperatures between 4.2 and 1.5 K using the technique described elsewhere.[4,11]

Plotted in Fig. 4 are the experimental G $(V)/G_N$ spectra obtained from a Sn/CrO_2 junction (point-contact No. 7), which were fitted using (a) the ballistic model and (b) the diffusive model. Displayed along with the spectra are the fitted values of P_c and Z. The data were collected at a temperature of approximately 1.75 K. We calculated a value of Δ for this temperature using the BCS approximation for Pb and Sn to get $\Delta=1.2$ and 0.59 meV, respectively, and kept them constant throughout the analysis. Also recorded was the experimentally determined value of $R_s \cong 0.75\ \Omega$, which was used to analyze the data, as all the contacts in this experiment were obtained in the same geometry. Using this procedure, both models gave nearly the same value of P_c ($P_b = 0.80 \pm 0.03$, $P_d = 0.82 \pm 0.03$). We want to emphasize here again that while the extracted values of P_c are the same, the two models give different values of Z. Spectra obtained with other junctions such as Pb/CrO_2 likewise gave P_c values for this sample of 0.84 as well but with different values of Z (see Fig. 5).

As mentioned previously, there should be no correlation between P_c and Z in the BTK formalism. We have also shown above that some of the P_c (Z) dependencies may be caused by systematic errors due to inconsistent analysis of the data. Nevertheless, we have observed such a correlation in at least some of the material systems, in which this correlation had been previously reported, most notoriously in CrO_2. To illustrate how Z affects P_c in our spectra, we plot P_c versus Z for our Pb/CrO_2 data in Fig. 5. We first use the ballistic formula, and obtain polarizations between 0.2 and

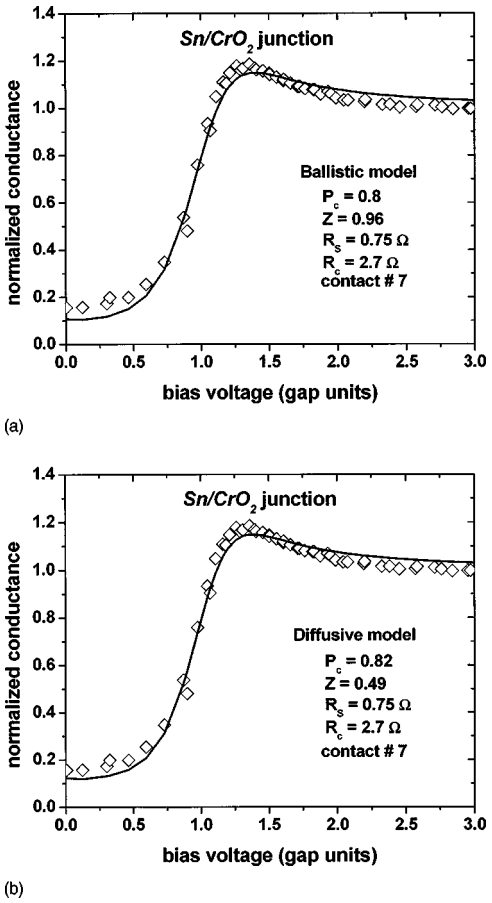

FIG. 4. Analyzed PCAR data of one point-contact spectrum of Sn/CrO_2 in the (a) ballistic limit and (b) the diffusive limit for positive bias voltage. The temperature, superconducting gap, and the spread-resistance used in the analysis was 1.75 K, 0.59 meV, and 0.75 Ω, respectively. (Note that the data points in the figure and all other analyzed curves have been corrected with R_s of the sample measured.) The negative bias voltage spectra were symmetric to the positive bias spectra in all cases.

0.83, with Z between 0.55 and 1.45. P_c indeed decreases with increasing Z in agreement with other studies of this material.[16] Despite the fact that there are no theoretical arguments for a linear relationship between P_c and Z, the fitted values in Fig. 5 show a fairly good linear dependence. However, if we extrapolate to $Z=0$ linearly, we obtain 1.13 ± 0.06, which is unphysical. In Ref. 16 a quadratic dependence of P_c (Z) for CrO_2 was proposed. While this is also hard to justify theoretically, a quadratic extrapolation gives P_c $(Z=0) = 1.05 \pm 0.29$. This result gives a more realistic number for P_c $(Z=0)$, closer to the theoretical value for this system,[21] but with a larger degree of uncertainty, which indicates that there are no statistical arguments for using a quadratic dependency for this set of data. This is, of course, related to the fact that we were not able to collect any data for this sample that could be described by the ballistic model with $Z<0.5$. On the other hand, the same spectra can be fitted by the diffusive model with practically the same polarization values, but with Z varying from 0 to 1.1. So, the diffusive model for $(Z \cong 0)$ yields $P_c = 0.84 \pm 0.03$ without any extrapolation.

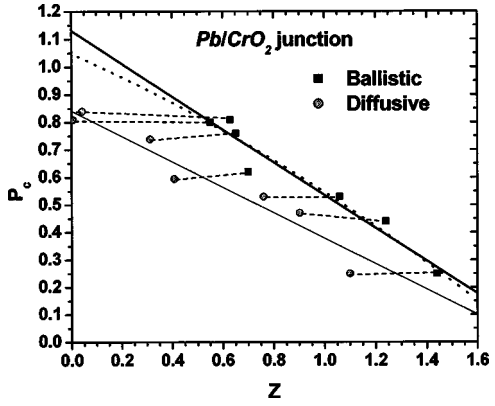

FIG. 5. P_c versus Z results obtained from analyzed point-contact spectra from a Pb/CrO_2 junction in the ballistic (filled squares) and the diffusive (empty circles). Extrapolations to $Z=0$ linearly (bold line) and quadratically (dotted line) give $P_c\ (Z=0)=1.13\pm0.06$ and $P_c\ (Z=0)=1.05\pm0.29$, respectively. The value for the diffusive model with no extrapolation yields $P_c\ (Z=0.042)=0.84\pm0.03$. The thin solid line traces the diffusive values. Dashed lines connect the two results from the same point contacts. The spread-resistance of this sample was $R_s\cong0.75\ \Omega$. The contact resistance values ranged from approximately 1 to 15 Ω.

Thus, if we were dealing with an unknown material we would have a dilemma: to either use the ballistic model and quadratic extrapolation to $Z=0$, but with a large uncertainty, or the diffusive model without extrapolation and thus with a smaller value of P_c (and, if the linear extrapolation would not yield $P_c>1.0$, we would have to think about this alternative as well). In this specific case, as the film was of relatively low quality and with high residual resistivity, it is likely that our sample does, indeed, have $P_c<1$. In other words, the correct value of P_c in this case is probably the one given by the diffusive model. The fact that we were not able to obtain any spectra with $Z_b<0.5$, which is the minimal Z_b that can be obtained in the diffusive regime (Fig. 1), can serve as a red flag suggesting that we are, indeed, in the diffusive regime. On the other hand, if in an experiment $Z_b \ll 0.5$ is observed, this is a good indication that ballistic formulas should be applied, with perhaps a subsequent extrapolation of some kind. As discussed earlier, if one fits an individual spectrum, P_c is practically the same for the two models but the values of Z are different. On the other hand, if one uses the ballistic formula and then extrapolates from $Z>0.5$ to $Z=0$, the resulting value for P_c would be quite different from the value found directly within the diffusive model for $Z\cong0$, and in fact incorrect and unphysical (larger than 1.0). From Fig. 5, we clearly see that the ballistic and diffusive estimates for P_c, corresponding to the same Z (but different contacts) are quite different. We conclude that if one measures a diffusive contact and then fits it with the ballistic formula, an extrapolation to $Z=0$ will be highly misleading. Unless one is confident that the measured contact is well within the ballistic regime, extreme caution should be exercised when using such extrapolation.

Let us now discuss a popular technique of extrapolating $P_c\ (Z)$ to $Z=0$, which is not without merits, in more detail. As stated above, there is no theory that suggests $P_c\ (Z)$ should be either linear or quadratic. However, Kant *et al.* (Ref. 18) proposed that P_c could be written as

$$P \approx P_0 \exp(-2\alpha\psi Z^2), \tag{2}$$

where P_0 is the intrinsic value of the spin-polarization, α is defined as the spin-flip scattering probability, and ψ is the ratio of the forward and backward scattering probabilities. The physical meaning of this formula, as opposed to the mathematical derivation which can be found in the original paper, is as follows: In the model of Ref. 18, the factor of Z^2 is derived from multiple scattering within the interface region (it is noteworthy that this assumption is applicable only for diffusive contacts, though the authors apply it in the ballistic case). Z^2 is thus proportional to the number of collisions and therefore to the ratio between the contact diameter and the electron mean-free path d/l. On the other hand, a natural (but not always correct) interpretation of the polarization suppression in the case of finite transparency is spin-flip scattering by defects at the interface. This is also proportional to the number of scattering events, albeit that only a small fraction of scattering will result in a spin flip. This immediately leads to Eq. (2), where $\alpha \ll 1$ is of the order of l/l_{sd}, where l_{sd} is the spin diffusion length. Interestingly, even when the actual data can be described by an exponential formula, the product $\alpha\psi$ both in Ref. 18 and our own similar calculations (see Table II) is of the order of, and not much smaller, than one. This simply reflects the fact that the assumption of a diffusive regime, implicitly used in the derivation, does not hold. On the other hand, it is obvious that for contacts with large Z and strong spin-flip scattering, the apparent value for P_c should tend to go to zero. Furthermore, since the total contact resistance R_N in the BTK model is proportional to $(1+Z^2)$, it is natural to assume that in many cases the spin-flip scattering, whether from impurities or not, depends on Z^2, and not on Z. Therefore, the exponential function, which smoothly interpolates between the two limits, may have some general validity. Nonetheless, there is no significant improvement in using Eq. (2) over a quadratic or even a linear dependence (compare the values of the χ^2 criterion for the three fits as shown in Table II. For all materials the three χ^2 values are very close, which indicates that all three extrapolations are of comparable statistical quality).

Finally, let us briefly touch upon another important issue: interface effects on superconductivity. So far we have only considered the case when the experimentally observed gap in the conductance spectra coincide with the bulk gap of the superconductor at a given temperature, and when the superconducting density of states is described by the BCS theory. This is the basis of both the original BTK model and its generalizations described above. However, in reality magnetic scattering of the Cooper pairs at and near the interface may be present, which would linearly suppress T_c and smear the density of states.[24] Within the fitting procedure this can be taken into account by substituting the measured temperature by a higher (effective) temperature.[25] Similar effects in the PCAR spectra may come from local heating in the contact area, which was discussed, for example, in Ref. 23. In the spectra used in this paper no substantial extra broadening was observed, proving that neither local heating nor interface

TABLE II. Fitted values from Eq. (1) for several ferromagnetic materials including CrO_2 from this work. Also included are statistical comparisons of the linear (χ_L^2), quadratic, (χ_Q^2), and exponential (χ_E^2) extrapolations.

Material	P_0	$\alpha\psi$	χ_L^2	χ_Q^2	χ_E^2
CrO_2[a]	0.93±0.03	0.245±0.05	35.6	45.1	36.5
CrO_2[b]	0.96±0.02	1.5±0.23	19.8	9.6	9.8
SRO[c]	0.58±0.01	0.59±0.2	1.8	5.2	1.9
SRO[d]	0.53±0.01	1.12±0.12	2.6	1.5	1.3
LSMO ($x=0.4$)[e]	0.82±0.02	0.31±0.03	10.8	6.8	7.7
LSMO ($x=0.3$)[e]	0.78±0.01	0.243±0.03	10.0	6.3	3.9
Ni[b]	0.38±0.01	1.94±0.18	1.1	2.0	0.9

[a]This work.
[b]Reference 16.
[c]Reference 22.
[d]Reference 17.
[e]Reference 10.

pair breaking was present in these contacts. However, in other experiments, broadening was observed, and sometimes so large that the effective temperature was higher than the bulk critical temperature,[26] implying that the pair-breaking effects, and not heating, were dominant in those particular contacts. However, in general it is difficult to distinguish between these two effects, since they affect the spectra in the same way, and, as it was pointed out in Ref. 18, there is no current theory to evaluate them independently.

In summary, we have discussed an analysis of PCAR spectra using the ballistic and diffusive models. By careful analysis of the PCAR data using this procedure, important information concerning the transport spin-polarization may be obtained on candidate materials for applications of spintronics devices. We have proven that both ballistic and diffusive models yield essentially the same values of the spin polarization (with the accuracy of approximately 3%) practically within the full range of P. We have also shown that in some cases the observed correlation between P_c and Z can be due solely to systematic errors in the data analysis. At the same time we have confirmed a previously observed correlation for P_c (Z) dependence in CrO_2, and some other material systems, in which case the interpolation to $Z=0$ is legitimate. At the same time we conclude that, as of now, there is no extrapolation formula that is significantly better than the others. We have also noted that if all available PCAR data correspond to a sizeable Z in the ballistic model, the ballistic conditions should be independently verified before extrapolating to $Z=0$. Much more work is needed to explain the mechanisms as to why the intrinsic value of the spin-polarization decreases when Z increases when analyzing PCAR spectra using either limit. Some of the assumption of the simple theory which models a point contact by an abrupt interface in one dimension with a step function voltage drop across a δ-function barrier can be legitimately questioned. It is encouraging, however, that, the modified BTK formalism seems to be able to absorb a number of physical effects well beyond the scope of the underlying model into a single number Z. Therefore, the values for the interfacial spin polarizations appear substantially more reliable than one could have anticipated from purely theoretical viewpoint.

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