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**DOI: 10.1063/1.5007623**

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## Diffusive-like effects and possible non trivial local topology on the half-Heusler YPdBi compound

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(Presented 8 November 2017; received 2 October 2017; accepted 21 November 2017; published online 11 January 2018)

The non-ambiguous experimental identification of topological states of matter is one of the main interesting problems regarding this new quantum state of matter. In particular, the half-Heusler family RMT (R = rare-earth, T = Pd, Pt or Au and T = Bi, Sb, Pb or Sn) could be a useful platform to explore these states due to their cubic symmetry and the topological properties tunable via their unit cell volume and/or the nuclear charges of the M and T atoms. In this work, we report electron spin resonance (ESR) and complementary macroscopic measurements in the Nd<sup>3+</sup>-doped putative topologically trivial semimetal YPdBi. Following the Nd<sup>3+</sup> ESR lineshape as a function of microwave power, size of the particle and temperature, we have been able to observe an evolution from a Dysonian lineshape to a diffusive-like lineshape. Furthermore, the Nd<sup>3+</sup> ESR intensity saturation is concentration dependent, which could be due to a phonon-bottleneck process. Comparing these results with the Nd<sup>3+</sup>-doped YPtBi, we discuss a possible scenario in which the Nd<sup>3+</sup> ions could locally tune the topological properties of the system. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5007623>

### I. INTRODUCTION

A new quantum state of matter,<sup>1</sup> the three dimensional topological insulators (3D TIs) has become a hot topic intensively explored by the community in order to understand their properties and the application of the topology in condensed matter physics.<sup>2,3</sup> The nontrivial insulators with gapless surface states, protected from backscattering and with highly spin polarized channels, have been the trigger to study a new field of topological systems. Although many experiments have been performed with different techniques,<sup>4,5</sup> the unambiguous experimental signatures of the topological phases are difficult to establish. There is a lack of experimental techniques to be compared with the most reliable probe to these states, which is angle resolved photoemission spectroscopy (ARPES).<sup>6–10</sup>

Previous theoretical predictions<sup>11</sup> claim that an impurity in a nontrivial matrix could bind to elementary quasiparticles excitations and, possibly, reflect locally the topology of the system. The half-Heusler intermetallic family RTBi (T = Pd, Pt) is a valuable platform to explore experimental hints of the nontrivial/trivial topology. Their cubic symmetry is an important symmetry to be explored in microscopic techniques such as electron spin resonance (ESR). Previous band structure calculations<sup>12–14</sup> show a topological phase transition (TPT) inside the family which are directly related with the subtle changes on the lattice parameter and the charges of the M and T atoms.

The local moment ESR (LMESR) lineshape is usually analyzed by the well accepted Dyson model.<sup>15–17</sup> The microwave electromagnetic field is attenuated on the surface of a metal due to its

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high conductivity. Thus, the microwave only penetrates a small length scale called skin depth  $\delta$ . In Dyson model, the LMESR is described in the diffusion-less regime, in other words, the diffusive time across the skin depth of the local moment  $T_D$  is negligible compared with the spin-spin transverse relaxation time  $T_2$ . In this limit, the lineshape is asymmetric with a ratio  $A/B \lesssim 2.6$  (defined in Fig. 2 a)) and is called Dysonian. In the case of an insulator, the size of the sample  $d$  is smaller compared with  $\delta$  and the lineshape is Lorentzian with  $A/B \approx 1$ . When  $T_D$  becomes comparable with the relaxation time  $T_2$ , which occurs in the conduction ESR (CESR),<sup>16</sup> Dyson model predicts the presence of diffusive effects, with  $A/B \gtrsim 2.6$ .

Recent ESR experiments in  $Y_{1-x}Nd_xPtBi$ <sup>18</sup> show a highly unusual diffusive-like ESR lineshape of the  $Nd^{3+}$  ( $g \approx 2.6$ ). The ESR lineshape of the  $Gd^{3+}$ , which is a natural impurity in the Y site, is Dysonian, while the  $Nd^{3+}$  resonance presents a completely diffusive lineshape. Varying the size of the particle, where  $\lambda = d/\delta$ , the microwave power, the concentration  $x$  and the temperature, the lineshape changes from Dysonian to diffusive. Interestingly, the ESR intensity saturates with different microwave power for different concentrations  $x$ . This is indicative of a phonon bottleneck regime. This result indicates that the phonon reservoir and the thermal bath are weakly coupled. The diffusive-like behavior could be connected with long lifetime phonons exciting highly mobile Dirac fermions within the skin depth via spin-orbit (SO) coupling. As a consequence, these Dirac fermions would diffuse over the skin depth.<sup>18</sup>

In order to verify this hypothesis in a counterpart compound in the same family, we report a systematic ESR study in  $Y_{1-x}Nd_xPdBi$ . The YPdBi is on the frontier of topological triviality and non-triviality<sup>12-14</sup> and could be a useful platform to contrast with the previous results reported for YPtBi. Recent crystalline electrical field (CEF) studies<sup>19,20</sup> show a systematic change in the sign of the crystal field parameters (CFP) comparing the YPdBi and the YPtBi compounds. This could be related with the inversion between the  $\Gamma_8$  (p-character) and  $\Gamma_6$  (s-character) bands, driven by the SO coupling. This inversion could change the charge distribution in the crystal structure, therefore reflecting in the CFP. Due to the fact that the YPdBi is on the frontier of trivial and nontrivial materials, those effects should be more pronounced in the YPdBi case, thus making a compelling case to be compared with YPtBi.

## II. EXPERIMENT

Single crystalline samples of  $Y_{1-x}Nd_xPdBi$  ( $0.005 \leq x \leq 0.1$ ) were synthesized using the Bi self flux technique. High purity elements with starting composition Y:Nd:Pd:Bi 1-x:x:1:20 were placed into an alumina crucible and sealed in an evacuated quartz tube. The samples were heated to 1170 °C for 4 hours then cooled down to 450 °C at 5 °C/h. The flux was removed using a centrifuge. The structure and phase purity of the studied samples were checked by x-ray powder diffraction using a commercial diffractometer (Cu-K $\alpha$ ). Elemental analysis were performed using Energy Dispersive Spectroscopy (EDS) and Wavelength Dispersive Spectroscopy (WDS). Magnetic susceptibility measurements were carried out in a SQUID magnetometer. Transport measurements were performed in a PPMS 9T. ESR measurements were performed for both single crystals and powdered samples in a X-band (9.5 GHz) spectrometer equipped with a goniometer and a He-flow cryostat. Different particle sizes and single crystals were used in this study. The studied samples masses ranged from 5 mg to 15 mg. Several different crystals from 5 batches were used.

## III. RESULTS AND DISCUSSION

Figure 1 a) shows the magnetic susceptibility as a function of temperature with  $H = 1.0$  T for  $Y_{1-x}Nd_xPdBi$ . The solid lines are the best fit of a Curie-Weiss law added to a Pauli temperature independent term. Assuming  $3.62 \mu_B/Nd$ , we estimate the concentrations  $x \approx 0.01, 0.03, 0.1$ . We considered only the range  $T \geq 150K$  in the fittings to avoid any CEF effects on this analysis.

In order to obtain the skin depth of these systems, we performed electrical resistivity as a function of temperature, shown in Fig. 1 b). At low temperature ( $T \lesssim 4$  K) it is possible to observe a small kink, probably associated with a possible superconductor phase in this system<sup>21</sup> or the presence of a binary superconductor. The skin depth is given by  $\delta = \sqrt{\rho/\pi f \mu}$  where  $f$  is the microwave frequency

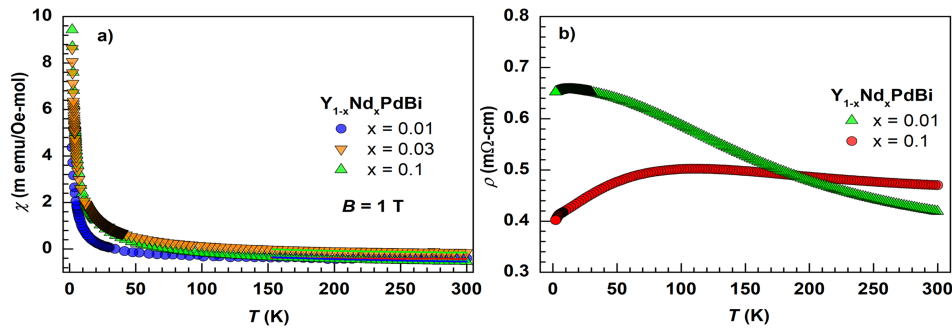


FIG. 1. a) Magnetic susceptibility as a function of temperature for  $Y_{1-x}Nd_xPdBi$  with  $H = 1.0$  T. b) Resistivity as a function of temperature for  $Y_{1-x}Nd_xPdBi$  with  $x \approx 0.01$  and  $0.1$ .

( $\approx 9.4$  GHz),  $\mu$  is the absolute magnetic permeability and  $\rho$  is the resistivity of the sample in a given temperature. Using  $\rho_{4K} = 0.6(1)$  m $\Omega$ -cm, we estimate  $\delta \approx 12(2)$   $\mu$ m, which is similar with the YPtBi skin depth.<sup>18</sup>

Fig. 2 a), b) and c) shows the ESR  $Nd^{3+}$ , which is associated to the  $\Gamma_6$  cubic CEF state, in  $Y_{1-x}Nd_xPdBi$ , for  $x = 0.1$ , as a function of the particle size. The hyperfine lines of  $Nd^{3+}$  in cubic symmetry with  $^{143}A = 213(5)$  Oe and  $^{145}A = 131(5)$  Oe split for the  $Nd^{3+}$  isotopes  $^{143}Nd^{3+}$  ( $I = 7/2$ ) and  $^{145}Nd^{3+}$  ( $I = 7/2$ ) were observed in the spectra (not shown).<sup>17,19</sup> Here we focus our analysis in the main ( $I = 0$ ) line. There is an evolution of the ratio  $A/B$  going from the smaller particle sizes (Fig. 2 a)) to the larger ones (Fig. 2 b) and c)). The lineshape of the  $Nd^{3+}$  resonance is a Dysonian for all the temperatures and microwave powers. The ratio  $A/B$  starts to decrease with high power due to saturation effects in the ESR lineshape, which could be associated to lineshape distortion in the saturated regime.<sup>17</sup>

The ratio  $A/B \lesssim 2.6$  for all the measurements of the ESR lineshape of the  $Nd^{3+}$  is an indicative that the YPdBi has an insulating character more pronounced than in the YPtBi compound, even though the two systems have nearly the same skin depth ( $\delta = 15$   $\mu$ m in YPtBi<sup>18</sup>). The  $Nd^{3+}$  g-value observed for those ESR were  $g = 2.641(1)$ , which is consistent with a  $\Gamma_6$  cubic CEF ground state.<sup>17</sup>

In order to look for diffusive effects, we have focused in the  $Nd^{3+}$  ESR in single crystals, which is shown in Fig. 3 for a)  $x = 0.01$ , b)  $x = 0.03$  and c)  $x = 0.1$ . In Fig. 3 a), which is for the  $Nd^{3+}$  ESR in

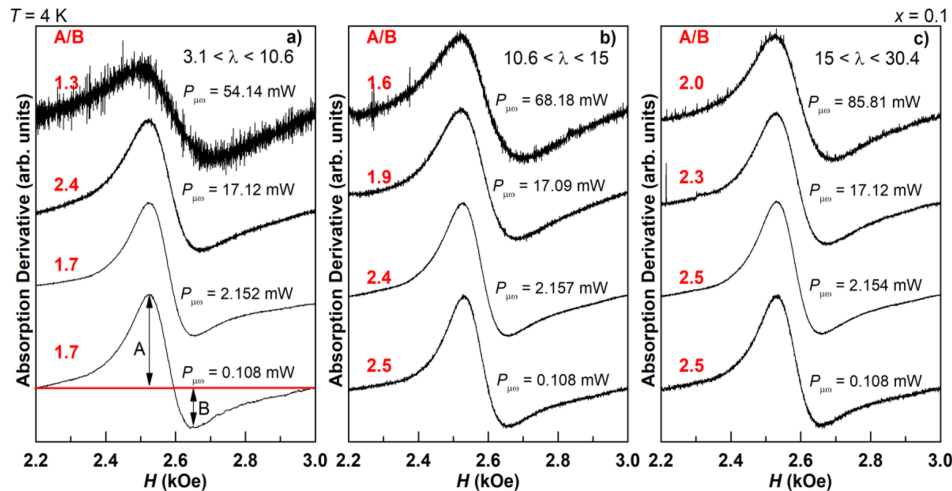


FIG. 2. Evolution of the ESR lineshape of the  $Nd^{3+}$  in  $Y_{0.9}Nd_{0.1}PdBi$  as a function of the microwave power for a)  $3.1 \leq \lambda \leq 10.6$ , b)  $10.6 \leq \lambda \leq 15$  and c)  $15 \leq \lambda \leq 30.4$ . In all cases the lineshape is Dysonian, even for high microwave power, within the saturation regime.

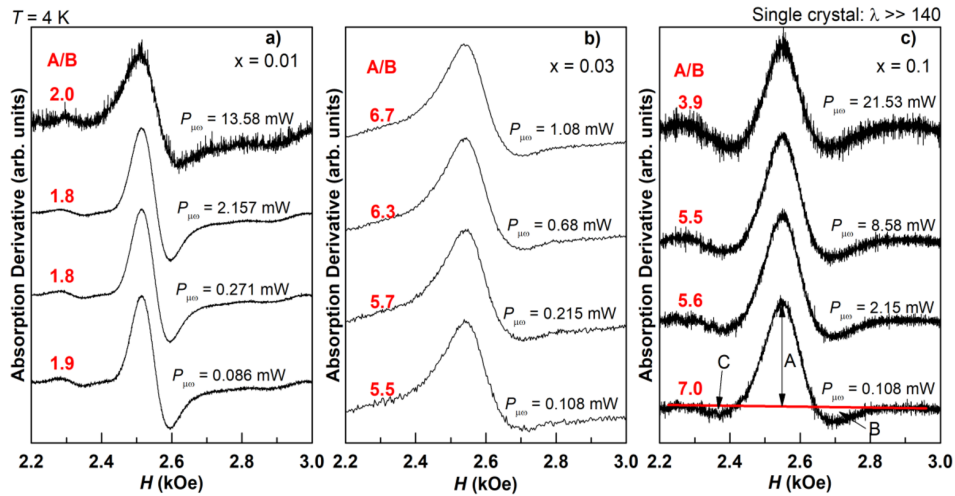


FIG. 3. Evolution of the ESR  $\text{Nd}^{3+}$  in  $\text{Y}_{1-x}\text{Nd}_x\text{PdBi}$  lineshape as a function of the microwave power for a)  $x = 0.01$ , b)  $x = 0.03$  and c)  $x = 0.1$ . All the experiments were done in single crystals. There is a clear evolution from a Dysonian lineshape ( $A/B \approx 1.9$ ), in the dilute case, to a diffusive-like lineshape in the more concentrated systems ( $A/B \approx 7.0$  for  $x = 0.1$ ).

the dilute regime, the ratio  $A/B$  obtained was  $\approx 1.9$ , which is characteristic of a Dysonian lineshape in a regime with the microwave skin depth somewhat smaller than the sample size. Interestingly, when the  $\text{Nd}^{3+}$  concentration is increased there is an evolution to a diffusive lineshape, which is unexpected for a LMESR. In Fig. 3 b), the ratio  $A/B$  increased to  $\approx 5.5$ , which means that the diffusive time  $T_D$  is becoming comparable to the relaxation time  $T_2$ .<sup>15-17</sup>

The most interestingly result is the  $\text{Nd}^{3+}$  resonance in single crystals with  $x = 0.1$ , which is shown in Fig. 3 c). The ratio  $A/B$  evolves to 7.0 and the appearance of a valley shaped curve with depth  $C$  appears on the left side of the resonance, characteristic of the diffusive effect in the ESR lineshape. Increasing the microwave power, it is possible to observe an increase of the value of  $C$ , but it is when the microwave power is saturating the  $\text{Nd}^{3+}$  ESR, which may cause some distortions in the ESR lineshape. Interestingly, all these results indicate that the diffusive character appears in very special conditions (high  $\text{Nd}^{3+}$  concentration, low temperature, higher microwave power and only in single crystals), which suggests that the phonon bottleneck regime, that has allowed the observation of diffusive effects in the lineshape of  $\text{Nd}^{3+}$  ESR in YPtBi, may be marginally relevant in YPdBi for these particular conditions.

Fig. 4 a) shows the  $\text{Nd}^{3+}$  ESR intensity of  $\text{Nd}^{3+}$  in  $\text{Y}_{1-x}\text{Nd}_x\text{PdBi}$  as a function of the microwave power for  $x = 0.01$  and  $x = 0.1$ . Similar results were obtained for  $x = 0.03$  (not shown). It is worth to notice that the  $\text{Nd}^{3+}$  ESR intensity for all concentrations saturates at reasonably low power ( $\approx 1.7$  mW) which indicates that the  $\text{Nd}^{3+}$  ions are in an insulating environment. For comparison, the microwave power which saturates the  $\text{Nd}^{3+}$  ESR in  $\text{Y}_{1-x}\text{Nd}_x\text{PtBi}$  is  $\approx 5$  mW.<sup>18</sup> This result corroborates with the first observation that the YPdBi system has a more insulating character than the YPtBi system, albeit the skin depth of the two systems are roughly the same. Furthermore, the microwave power necessary to saturate the  $\text{Nd}^{3+}$  ESR in YPdBi at  $T = 4$  K is  $x$ -dependent, which indicates a phonon bottleneck process in this system.

To further understand the existence of a phonon bottleneck effect in the  $\text{Nd}^{3+}$  ESR in YPdBi, we have investigated, in greater details, the temperature and microwave power dependence of the diffusive effects in the  $\text{Nd}^{3+}$  ESR in YPdBi. Fig. 4 b) shows the evolution of the ratio  $A/B$  for the  $\text{Nd}^{3+}$  ESR in  $\text{Y}_{0.97}\text{Nd}_{0.03}\text{PdBi}$  single crystals with  $P = 0.54$  mW. Although it is not possible to observe any traces of a  $C$  on the left side of the main  $\text{Nd}^{3+}$  ( $I = 0$ ) line, the ratio  $A/B$  at 4 K is 6.2 (diffusive) and evolves to 2.7 (Dysonian) at 21 K.

As Fig. 3 c) indicates, the diffusive effect is more pronounced in the more concentrated samples. Fig. 4 c) shows the microwave power dependence of the ratios  $A/B$  and  $C/B$  for the ESR of  $\text{Nd}^{3+}$  in  $\text{Y}_{0.9}\text{Nd}_{0.1}\text{PdBi}$  single crystals at 4 K. The ratios  $A/B$  and  $C/B$  evolves systematically

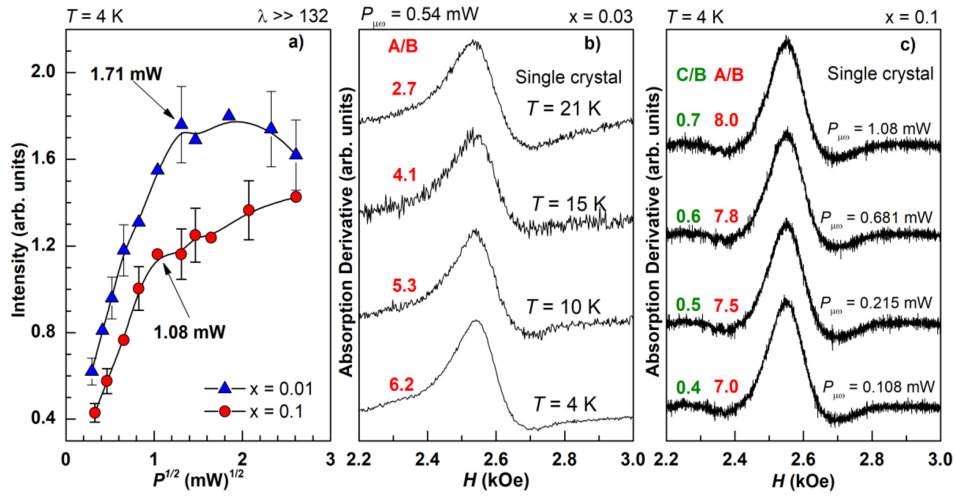


FIG. 4. a) ESR intensity of  $\text{Nd}^{3+}$  in  $\text{Y}_{1-x}\text{Nd}_x\text{PdBi}$  as a function of the microwave power for  $x = 0.01$  and  $x = 0.1$ . b) Evolution of the ESR lineshape of  $\text{Nd}^{3+}$  in  $\text{Y}_{0.97}\text{Nd}_{0.03}\text{PdBi}$  as a function of the temperature. c) Evolution of the ESR lineshape of  $\text{Nd}^{3+}$  in  $\text{Y}_{0.9}\text{Nd}_{0.1}\text{PdBi}$  as a function of the microwave power.

with the microwave power until the system is saturated, which is the same behavior observed for  $\text{Y}_{1-x}\text{Nd}_x\text{PtBi}$ .<sup>18</sup> These two results (the evolution with temperature and with the microwave power) strongly suggests that the origin of the diffusive effects in the  $\text{Nd}^{3+}$  ESR is the same in  $\text{YPdBi}$  and  $\text{YPtBi}$ , but the diffusive effects are much more subtle in the former.

In fact, the Dysonian  $\text{Nd}^{3+}$  ESR lineshape for the dilute samples and the evolution of the  $\text{Nd}^{3+}$  ESR lineshape as a function of the particle size assures us that the  $\text{Nd}^{3+}$  local moments are probing the presence of  $ce$  within the skin depth. However, it has been shown that the spin-lattice relaxation from  $\text{Nd}^{3+}$  and  $\text{Er}^{3+}$  doped-samples via  $ce$  is very weak in  $\text{YPdBi}$ .<sup>20</sup> Therefore, the relaxation to the thermal bath should be mainly via phonons by the SO coupling. It is important to note that the  $\text{Nd}^{3+}$  ESR saturates very rapidly for  $P \approx 2$  mW, which indicates an insulator environment for  $\text{Nd}^{3+}$ . All those facts assure us that insulating and metallic behavior coexists within the skin depth in  $\text{YPdBi}$ .

The most strikingly result obtained in this study in comparison to those reported for  $\text{YPtBi}$ <sup>18</sup> is that the diffusive-like effect in  $\text{YPdBi}$  only appears for the ESR measurements in single crystals, which means that is more restrict to a small range of larger  $\lambda$ . Furthermore, the temperature, microwave power, sample size and  $\text{Nd}^{3+}$ -concentration dependence of the diffusive effects in  $\text{Nd}^{3+}$  ESR in  $\text{YPdBi}$  are strong evidences that this effect is not solely a simple result of the phonon bottleneck process that occurs in this system. In fact, the presence of some fraction of highly mobile Dirac electrons on the surface of this materials may be required to understand our results in comparison to those reported for  $\text{YPtBi}$ .<sup>18</sup> Although a trivial system, the  $\text{YPdBi}$  is on the frontier of the topological non-triviality according to previous band structure calculations<sup>12-14</sup> and any change in the atoms could tune locally or globally the topology of the system.

The diffusive-like effect observed in the  $\text{Nd}^{3+}$  resonance in  $\text{Y}_{1-x}\text{Nd}_x\text{PdBi}$  occurs only in extreme conditions (low temperatures, high concentration, larger sample sizes and higher microwave power) compared with  $\text{YPtBi}$ ,<sup>18</sup> thus it seems the  $\text{Nd}^{3+}$  ions in  $\text{YPdBi}$  could be locally tuning the topology of the system. This hypothesis corroborates with the fact that the diffusive-like effect observed in the  $\text{YPtBi}$  compound appeared in a wide range of condition, indicating that the non-trivial topology and the presence of Dirac electrons is a global effect, which does not occurs in  $\text{YPdBi}$ . As such, the enhancement of the SO coupling introduced by  $\text{Nd}^{3+}$  substitution and/or a change of the carrier contribution near the Fermi level, which alters the CEF effects of the system,<sup>20</sup> could cause a band inversion around the local moment and, as a consequence, there might be a local transition from trivial to nontrivial topology in  $\text{YPdBi}$ . Further investigations for other compounds within the half-Heusler family would be valuable to continue to investigate the interplay between topology and ESR diffusive effects.

#### IV. CONCLUSION

In summary, we were able to show the existence of a diffusive-like effect on the  $\text{Nd}^{3+}$  ESR in  $\text{Y}_{1-x}\text{Nd}_x\text{PdBi}$ . We have found that, although the ESR signal is from a local moment ( $\text{Nd}^{3+}$ ), it is possible to observe a diffusive effect in particular conditions - low temperatures, high  $\text{Nd}^{3+}$  -concentrations, larger sample sizes (single crystals) and higher microwave power. The diffusive-like effects on the  $\text{Nd}^{3+}$  ESR in  $\text{YPdBi}$  have revealed temperature and microwave power dependencies, which is highly unusual and cannot be explained solely by a phonon bottleneck regime. We argue that it could be a manifestation of a local topological phase transition caused by the  $\text{Nd}^{3+}$  ions. These ions introduce small changes on the lattice parameters that could be enough to slightly modify the charge distribution on the residual Fermi surface, which affects the CEF effects in these materials. Additionally, the  $\text{Nd}^{3+}$  -substitution in  $\text{YPdBi}$  can enhance the SO coupling locally and, along with the CEF effects, can induce a transition to a non-trivial topology around the impurity. Further investigations would be valuable to understand the interplay between topology and ESR diffusive-like effects observed in the compounds of the half-Heusler family.

#### ACKNOWLEDGMENTS

This work was supported by FAPESP (SP-Brazil) Grants No 2013/17427-7, No 2012/04870-7, 2006/60440-0, 2009/09247-3, 2010/11949-3, 2011/01564-0, 2011/23650-5, 2012/05903-6, CNPq Grants No 442230/2014-1 and 304649/2013-9, CAPES and FINEP-Brazil. The authors acknowledge the Brazilian Nanotechnology National Laboratory (LNNano) for providing the equipment and technical support for the EDS experiments.

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