

Reply to “Comment on ‘Nature and entropy content of the ordering transitions in $R\text{Co}_2$ ’”

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In the previous Comment, Forker and co-workers claim that perturbed angular correlation (PAC) data leave no alternative to the conclusion that the spontaneous magnetization of PrCo_2 and NdCo_2 undergoes a discontinuous, first-order phase transition at T_C . We show here that their claim is in clear contradiction with a wealth of experimental evidence, including our own. Finally, we propose a possible origin for the disagreement between their interpretation of the PAC results and the literature on this subject.

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I. INTRODUCTION

Before the publication by Forker *et al.* of “Perturbed angular correlation study of the magnetic phase transitions in the rare-earth cobalt Laves phases $R\text{Co}_2$ ” in 2003,¹ there was a general consensus in the literature on the nature of the magnetic ordering phase transitions of the $R\text{Co}_2$ series. With a rather extensive experimental base, the magnetic ordering transitions in the Co Laves phases with $R=\text{Er}$, Ho , and Dy were always classified as first-order transitions (FOTs) and the rest as second-order transitions (SOTs). Initially there had been some controversy as to the reason why in NdCo_2 and PrCo_2 magnetic ordering was not a FOT, as the available models of the $R\text{Co}_2$ series^{2,3} indicated. Apparently, the riddle had been clarified^{4,5} by introducing the effect of cell volume on the series expansion of the free energy.

However, a series of ¹¹¹Cd perturbed angular correlation (PAC) experiments led Forker *et al.* to partially review the literature on this subject, concluding that the “few detailed experimental studies of the phase transitions in the light $R\text{Co}_2$ ” available leave some doubts “as to the classification of the transitions of NdCo_2 and PrCo_2 as SOTs.”

The accumulation of experimental results suggesting the SOT character of NdCo_2 and PrCo_2 transitions is in our opinion rather conclusive, but one might be forced to admit that several of those *pieces of evidence* in the literature are reached by *inspection* of temperature-dependent experimental data, and some of them may be not as conclusive as would be desirable. In the opinion of Forker *et al.*,¹ the clear doubts about the SOT character of the NdCo_2 and PrCo_2 Curie transitions are fully solved by PAC measurements. As they expose again in their previous Comment, in their opinion PAC leaves no alternative to the conclusion that the spontaneous magnetization of PrCo_2 and NdCo_2 undergoes a discontinuous, first-order phase transition at T_C .

This forceful conclusion by Forker *et al.* as well as the large magnetocaloric effect of $R\text{Co}_2$ FOTs were the motivations to perform our differential scanning calorimetry (DSC) study of $R\text{Co}_2$.^{6–8} As it is known, DSC is well suited to determine the thermodynamic character of a phase transition because proper integration of the calorimetric signal yields

the latent heat in FOTs while in a SOT, the signal reflects a continuous change in entropy. Moreover, the application of a magnetic field also helps to discriminate SOTs from FOTs by field-dependent DSC measurements. Our experimental results indicating a SOT character for NdCo_2 and PrCo_2 were clearly in disagreement with the conclusion of Forker *et al.*, which is stated again in their previous Comment.

This reply is organized as follows: in Sec. II we will show that the claim of Forker *et al.* is in clear contradiction with a wealth of experimental evidence in the literature. In Sec. III, we analyze the dependence of the critical temperature on the magnetic field and we apply the Banerjee criterion to $R\text{Co}_2$ magnetization data ($R=\text{Nd}, \text{Pr}, \text{Er}, \text{Ho}$). Finally, in Sec. IV we propose a possible origin for the disagreement between the interpretation of the PAC results and the literature on this subject. Finally, in Sec. V, we summarize our results.

II. PREVIOUS RESULTS

To clarify the present state of the subject, it is relevant to review some of the previous literature on the subject and the view of Forker *et al.* on it.

Specific heat. Of particular importance are the adiabatic calorimetry data of NdCo_2 and PrCo_2 published by Deenadas *et al.*⁹ in 1972. Specific heat measurement is a crucial experiment on this subject, although the work passed unnoticed by Forker *et al.* in both Ref. 1 and their previous Comment. To state it simply, the C_p curves are as incompatible with a FOT character for NdCo_2 and PrCo_2 Curie transitions as our DSC results^{6,7} are. One would argue about these measurements (as the previous Comment does on our DSC) that the prototypical SOT shape of NdCo_2 and PrCo_2 specific heat curves at T_C may be caused by a (rather capricious) distribution of inhomogeneous phases with critical temperatures that *obfuscate*¹⁰ the FOT-peaked shape. But this strange phenomenon is shown false by one of the samples itself: NdCo_2 offers a first-order spin reorientation transition (SRT) just ~ 60 K below the magnetic ordering transition as a perfect *witness* on how much a FOT is affected by sample quality *on the very same specimen*. Both NdCo_2 samples, the one

measured in Ref. 9 and our own,⁶ show a very abrupt, clearly first-order peak at $T_{SRT}=40$ K. The full width at half maximum (FWHM) of the FOT peaks are <1 K and 0.5 K in Deenadas *et al.* and our DSC, respectively, ruling out the hypothesis about a broad distribution of inhomogeneous phases with different critical temperatures in our samples. Of course there is some distribution in the Curie temperature, but as shown experimentally, it is not as broad as to fully obfuscate a FOT peak. In contrast, a nicely broad, λ -shaped, SOT-like peak is observed at the magnetic ordering, which spreads as much as several decades around the Curie temperature. Indeed, the FWHM of the λ peak at T_C is ~ 15 times larger than the FWHM at T_{SRT} in both works. Although PrCo_2 does not offer such a witness peak, the shape of the specific heat and DSC curves at the order transition is hard to reconcile with a FOT character.

Besides, those arguments given by calorimetry are not the only sound ones pointing in the same sense.

Transport properties. NdCo_2 and PrCo_2 do not present the abrupt drop in resistivity shown in the Er, Ho, and Dy Laves Co phases, as clearly shown by several studies: see, for example, the recent reviews by Duc and Brommer¹¹ and Gratz and Markosyan,¹² but above all those by Hauser *et al.*,¹³ Deenadas *et al.*,⁹ and Duc *et al.*,⁴ which in our opinion can only be interpreted as a clear evidence of a SOT in PrCo_2 and NdCo_2 at the Curie temperature. Indeed the authors of the cited works do identify PrCo_2 and NdCo_2 magnetic orderings as SOTs in all those cases, contrary to the reinterpretation of Forker *et al.* of their works.

Mössbauer spectroscopy. There are also temperature-dependent Mössbauer experiments¹⁴ on NdCo_2 , which are of special interest here, as PAC is also a hyperfine technique. Unfortunately, the number of spectra shown in Ref. 14 is scarce (the temperature step is of the order of 10 K) and therefore it is difficult to conclude on the order of the transition. But contrary to the point of view expressed by Forker *et al.* (as if the results were positive to a FOT) the fact that Atzmony *et al.* do measure in NdCo_2 one spectra in the middle of the transition (attaining about 50% of the saturation signal in the vicinity of T_C) is much more likely for a SOT than for a FOT.

Structure and magnetostriction. We might agree with Forker *et al.* that a FOT is not excluded (as it is not a SOT, either) by some of the studies of the temperature dependence of the lattice parameters and the crystal distortions¹⁵ previous to the publication of Ref. 1. More recently, Ouyang *et al.*¹⁶ present a very detailed temperature-dependent study showing the continuous character of the changes of the lattice constants at the Curie temperature, in strong contrast with those taking place at T_{SRT} . Ouyang and co-workers also show NdCo_2 anisotropic magnetostriction data, with similar characteristics: a FOT is hard to reconcile with their data at T_C , as the anisotropic magnetostriction constant λ_{100} presents a discontinuous drop at T_{SRT} which is absent at T_C . Indeed, a previous neutron scattering work by Lee and Pourarian¹⁷ already was very clear about the SOT character of NdCo_2 and PrCo_2 as the thermal and magnetic strains were very similar to those of Tb and very different to those of Dy, Er, and Ho. We do not agree with Forker *et al.* in their comments about the implications of Lee and Pourarian's work the current subject.

Theory. The first models on the critical behavior of the $R\text{Co}_2$ compounds,^{2,3} which assumed a rigid structure of the Co band through the series, suggested that the low T_C of NdCo_2 and PrCo_2 should correspond to a FOT. However, later calculations showed that the lanthanide contraction along the series drastically changes the mechanism of the Co-moment formation at T_C from light to heavy rare-earth $R\text{Co}_2$ compounds. Khmelevskiy and Mohn⁵ are as clear as follows: “a phase transition in the light-rare-earth compounds PrCo_2 , NdCo_2 [...] is impossible because the Itinerant Electron Metamagnetism conditions are not fulfilled for the d subsystem and the Co atoms carry a magnetic moment caused by spontaneous polarization due to the exchange interaction within the Co d band. The second-order transition in these compounds is thus a consequence of the internal properties of the d subsystem [...]”

However, the interpretation of the PAC data on NdCo_2 and PrCo_2 presented in Ref. 1 is that *there is no way to avoid the conclusion* that in PrCo_2 and NdCo_2 the spontaneous magnetization undergoes a FOT at T_C . Given the disagreement of PAC results with the previous literature, it would really be of interest to find some clear criteria to solve the riddle.

III. FOT-SOT DISCRIMINATION CRITERIA

T_C field dependence. A typical behavior of magnetic FOTs is the dependence of T_C with the applied field. The shift of the $T_C(H)$ has been clearly observed in many systems showing magnetic FOTs, including MnAs,¹⁸ $\text{Gd}_5\text{Si}_{4-x}\text{Ge}_x$,¹⁹ manganites,²⁰ pyrochlores,²¹ and $R\text{Co}_2$ ($R=\text{Er}$, Dy, and Ho),^{22,23} among others.

This behavior is indeed directly connected with the FOT character of the magnetic transition, as first pointed out by Meyer and Taglang.²⁴ During the 1960s a general theory of magnetic ordering FOTs was developed,^{18,25,26} including different mechanisms leading to first-order transitions. All those works coincide to predict that as soon as the Curie transition is a SOT, T_C does not depend on the applied field, while a $\partial T_C/\partial H \neq 0$ is predicted in the FOT case.

In Fig. 1 we show the shift of the critical temperature [$\Delta T_C(H)=T_C(H)-T_C(H=0)$] as a function of applied magnetic field measured on the $R\text{Co}_2$ series. The ΔT_C for $R=\text{Ho}$ (■), Er (●), Nd (▽), and Pr (□) have been obtained from our DSC measurements,⁷ while DyCo_2 (⊙) data are taken from magnetostriction measurements by del Moral and Melville,²³ which expands to much higher fields than our DSC data on DyCo_2 . The straight lines are linear fits to the observed values. The following slopes are obtained: 2.0 ± 0.1 K/T for ErCo_2 , 3.7 ± 0.1 K/T for HoCo_2 , 9.1 ± 0.1 K/T for DyCo_2 , -0.04 ± 0.08 K/T for PrCo_2 , and 0.06 ± 0.08 K/T for NdCo_2 . Clearly, the PrCo_2 and NdCo_2 values are compatible with a constant T_C , as a visual inspection of Fig. 1 suggests. The spin reorientation temperature of NdCo_2 shows a very well-defined linear dependence of T_C with the magnetic field, with slope $\partial T_{SRT}/\partial H = 3.1 \pm 0.3$ K/T, as shown in the inset to Fig. 1. In our opinion, the contrast between $\partial T_{SRT}/\partial H$ and $\partial T_C/\partial H=0$, obtained in a single measurement series on the same sample, and the

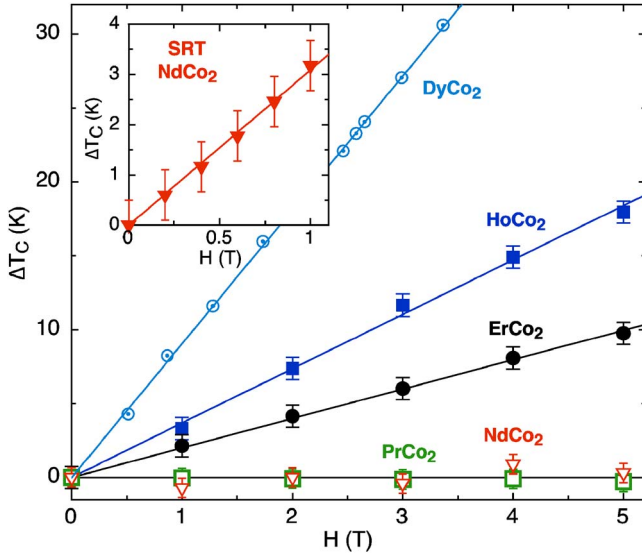


FIG. 1. (Color online) Variation of the critical temperature with the applied magnetic field in the $R\text{Co}_2$ series. The inset shows data for the spin reorientation transition in NdCo_2 .

different shapes of the specific heat and DSC curves at T_{SRT} and T_C are extremely strong arguments in favor of a FOT SRT and a SOT magnetic ordering in NdCo_2 and, by extension, in PrCo_2 .

Banerjee criterion. In 1964, Banerjee²⁷ condensed Landau-Lifshitz^{28–31} and Bean-Rodbell¹⁸ criteria providing a tool to distinguish magnetic FOTs from SOTs by purely magnetic methods³²; the presence or absence of a negative slope region on the isotherm plots of H/M vs M^2 near the critical temperature indicates a FOT or a SOT transition, respectively. Recently, this criterion has been applied with success to several systems.^{20,21,33} In fact, Banerjee criterion is equivalent to test the S shape of an Arrott plot (M^2 vs H/M), which has already been used as a test for magnetic FOTs in $R\text{Co}_2$ with heavy rare-earth elements.¹¹

In Fig. 2, we show the H/M vs M^2 plot of isotherms of NdCo_2 (\blacklozenge), PrCo_2 (\square), ErCo_2 (\bullet), and HoCo_2 (\blacksquare). The magnetization of ErCo_2 and HoCo_2 has been measured at $T=37$ K and $T=84$ K, i.e., ~ 4 K above T_C in order to evidence the effect of the field-induced FOT at about $H=1$ T. The magnetization curves of NdCo_2 and PrCo_2 have been measured at their corresponding T_C ($T=41.6$ K for PrCo_2 and $T=100$ K for NdCo_2), as it is clear from Fig. 1 that a field $H \leq 5$ T would not induce the transition at a higher temperature.

The same samples as those used in Refs. 7, 8, and 34 have been used, and their detailed structural and magnetic characterization has already been given. From Fig. 2 it is evident that ErCo_2 and HoCo_2 fulfill the Banerjee criterion for FOTs, as the negative-slope region coincides with the field-induced transition. PrCo_2 and NdCo_2 present an homogeneously increasing slope along the whole curve. Therefore, the magnetic ordering transition of PrCo_2 and NdCo_2 must be identified as a SOT. A complete study of the Banerjee criterion applied to $R\text{Co}_2$, including magnetization measurements at different temperatures and fields above and below the critical

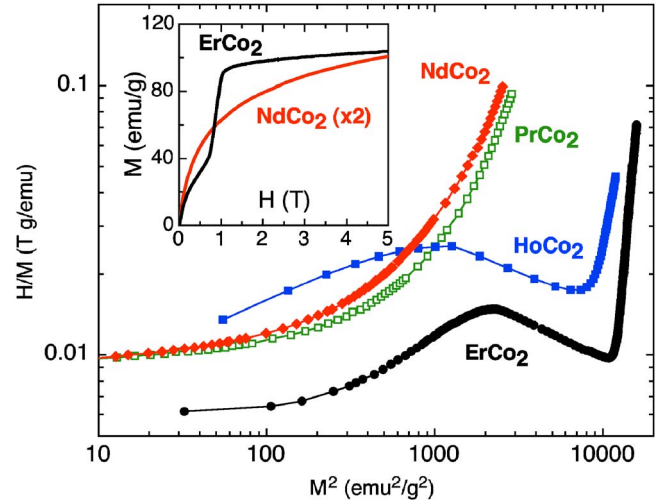


FIG. 2. (Color online) Double-log H/M vs M^2 plot of isotherms of NdCo_2 , PrCo_2 , ErCo_2 , and HoCo_2 measured at temperatures slightly above $T_C^{H=0}$. Only the FOTs show a negative-slope section. The inset shows the magnetization curves of ErCo_2 and NdCo_2 (which has been multiplied by a factor of 2 for clarity).

temperature, is in progress and will be published elsewhere. The magnetization curves of ErCo_2 and NdCo_2 are shown for comparison as an inset to Fig. 2. NdCo_2 data have been multiplied in the inset by a factor of 2 for clarity.

IV. XMCD MEASUREMENTS AND THE $R\text{Co}_2$ ELECTRONIC STRUCTURE

$R\text{Co}_2$ are systems with a complex electronic structure. This electronic complexity may be at the origin of the controversy between Forker’s interpretation of PAC measurements and every other significant piece of experimental work on NdCo_2 and PrCo_2 magnetic transition known to us.

Probably, the pivotal role in the subject we are dealing with is played by the fact that PAC measurements sample a very particular component of the $R\text{Co}_2$ magnetization, as the dominant contribution to the ^{111}Cd hyperfine field comes from the s -electron spin polarization, as explained in the previous Comment.¹⁰ Forker *et al.* assume the following hypothesis: *the s -electron spin polarization is induced by (and proportional to) the polarized $3d$ -band*, and their interpretation is based on this fact. They assume that a PAC measure of the s -band magnetic polarization is proportional to the Co $3d$ moment.

We have performed x-ray magnetic circular dichroism (XMCD) experiments on ErCo_2 . The XMCD spectra have been recorded at the Co $L_{2,3}$ and K edges (corresponding to Co $2p \rightarrow 3d$ and Co $1s \rightarrow 4p$ transitions, respectively) and at the $M_{4,5}$ Er edges (corresponding to Er $3d \rightarrow 4f$ transitions). The experiments have been performed at 4.0.2 beamline at ALS and ID08 and ID12 beamlines at ESRF. The experimental details are given elsewhere.^{34–36}

Although it is well established that $L_{2,3}$ and $M_{4,5}$ edges probe the $3d$ Co and the $4f$ Er magnetization, respectively, what is observed in the K Co edge is the polarization of the Co sp band,^{37,38} which is strongly hybridized with the $5d$

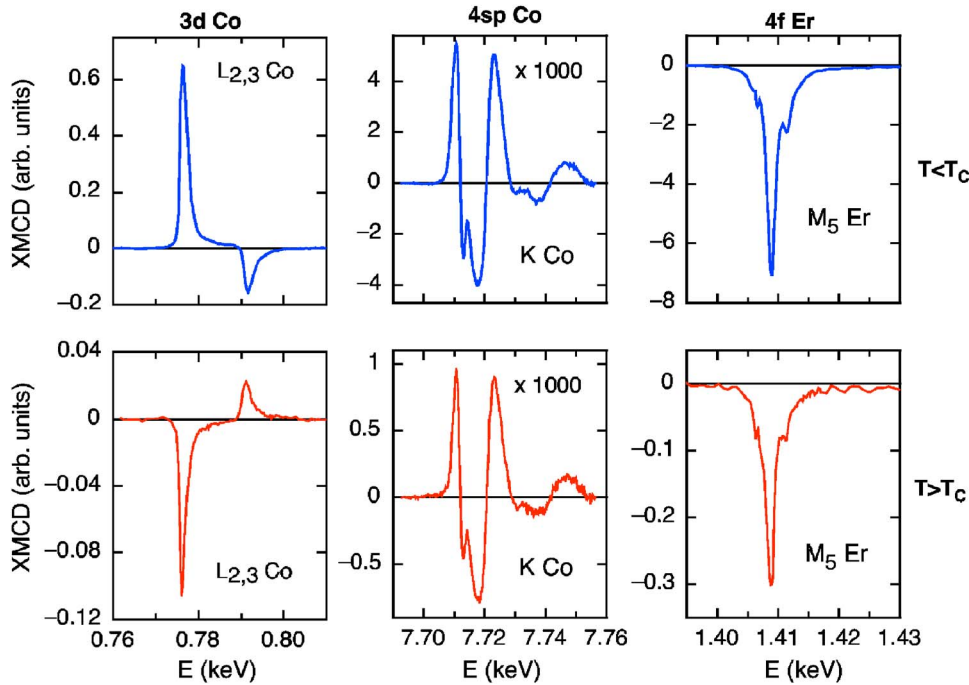


FIG. 3. (Color online) XMCD spectra recorded on ErCo_2 at the Co $L_{2,3}$ edges (left panels), Co K edge (central panels), and Er M_5 edge (right panels). The XMCD spectra have been obtained at 5 K (upper panels) and 90 K (lower panels) under an applied field of 1 T.

rare-earth band.^{35,36,39} In particular, in $R\text{Co}_2$ the effect of the rare-earth moments on the Co sp polarization is very strong, as our dichroic measurements show. In Fig. 3 we compare the dichroic spectra obtained at 90 K (well above T_C , lower panels) and at 5 K (well below T_C , upper panels), under an applied field of 1 T, at the Co $L_{2,3}$ edge (left panels), the Co K edge (central panels), and the Er M_5 edge (right panels).

First we focus in the fact that Er M_5 and Co $L_{2,3}$ edges, thus Er $4f$ and Co $3d$ electrons, respectively, show the expected behavior of a ferrimagnet under a moderate applied field, as indeed ErCo_2 is. The magnetization of the sublattice with the largest magnetic moment (Er) is, on average, always parallel to the applied magnetic field, thus maintaining its sign unaffected by the phase transition. Note that, as expected, the Er M_5 XMCD magnitude is strongly enhanced, as the magnetization is. The magnetization of the Co $3d$ band is probed by the XMCD measured at the Co $L_{2,3}$ edges.

The Co $3d$ magnetization, with a much smaller magnetic moment than the Er ions, changes its sign at low temperature, as expected in a ferrimagnet. Surprisingly enough, the polarization of the Co sp band, probed by the Co K -edge XMCD, does not behave in a proportional manner to the Co $3d$ moments. As the two central panels of Fig. 3 show, the magnetic moment induced in the Co sp band *does not* change sign upon the ferrimagnetic transition. The sp Co magnetization has the same sign both above and below T_C , just as the $4f$ rare-earth magnetic moment. Moreover, the shape of the measured K -edge spectra is strongly different to the Co metal one. This clearly shows that the sp Co band is strongly hybridized with the rare-earth $5d$ band and it is much more influenced by the $4f$ moment than by the Co $3d$ one, as it has been suggested in the recent literature.^{35,36,39,40} This fact offers a clue to understand the apparent disagreement of the PAC results with ours^{6,7} and the rest of the experimental results already cited about the nature of the NdCo_2 and PrCo_2 transition. Two facts may thus be relevant.

(i) As shown by our XMCD results, the magnetic polarization of the sp band is not proportional to the polarization of the Co $3d$ band, but it is strongly influenced by the rare-earth moments. This has been well established by K -edge XMCD measurements in rare-earth intermetallics, and it rules out the base hypothesis for PAC interpretation by Forker *et al.*

(ii) Moreover, the PAC spectrum is a local probe and ^{111}Cd occupies the rare-earth site in $R\text{Co}_2$. Therefore, as the polarization of the Co sp band is strongly influenced by the $4f$ rare-earth moments (via the $5d$ band), one would expect that the local polarization of the sp band is strongly affected by the absence of the rare-earth ion at the probing site. If this is the case, ^{111}Cd PAC in $R\text{Co}_2$ would give a perturbed signal, which may not be easy to correlate with the sp polarization of the parent undoped material. However, PAC is not our field of expertise and we leave this discussion open to the hyperfine-probe community.

V. CONCLUSIONS

We have shown that the SOT character of the NdCo_2 and PrCo_2 transitions from paramagnetism to ferromagnetism is established out of any reasonable doubt, except for the fact that the temperature evolution of the PAC spectra remains unexplained. It is clear from the literature that PAC spectroscopy has been used with success to study phase transitions (see references cited in Refs. 1 and 10) in other systems. We do not doubt the general validity of the technique, but $R\text{Co}_2$ is probably a not very favorable system, due to the complexity of its electronic structure (at the edge of Co moment formation) and to the fact that ^{111}Cd PAC in $R\text{Co}_2$ occupies the R site, locally affecting the Co sp moment in a very strong way.

We deal here with the interpretation of a series of PAC results, which is in disagreement with every other relevant results published on the issue. In our opinion, this gives rise to a problem for the interpretation of the PAC data on this particular subject, and it is not justified to take it as a proof of its unique capabilities to tackle the subject. This positioning disregards every previous piece of work on the FOT vs SOT classification of the NdCo₂ and PrCo₂ Curie transitions.

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