

Magnetic and structural ordering in SmH_{2+x} through electrical resistivity measurements

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Measurements of the electrical resistivity in SmH_{2+x} hydrides ($x \leq 0.26$) show that there exists a structural ordering at low temperature as soon as $x > 0$, i.e., it occurs within the H atoms occupying the octahedral interstitial sites. It is thus possible to define at low temperatures a relaxed R and a quenched Q state. For $x \leq 0.1$, we observe a sharp magnetic transition at $T_N = 9.6$ K which smooths at higher x values and disappears for $x = 0.26$. The origin of this disappearance is probably related to crystal-field splitting effects which are apparently very weak for x close to 0. We observe also a resistivity minimum just above T_N for $x > 0$ (whose amplitude is always somewhat larger in the Q state), which is probably related to the occurrence of an antiferromagnetic incommensurate ordering or to corresponding critical fluctuations.

INTRODUCTION

The magnetic and structural properties of SmH_{2+x} compounds have been investigated through measurements of the specific heat,¹ the magnetic susceptibility,¹⁻³ and the Knight shift,^{3,4} by structural studies via neutron⁵ and x-ray diffraction⁶ as well as NMR.⁷ The analysis of these data does not lead to a completely coherent understanding of the electronic and atomic aspects of these materials. As concerns, for instance, the magnetic properties, one of the important features is the influence of the crystal-field effects (CFE); *a priori*, one expects a situation very similar to the cerium hydrides (because Sm and Ce have the same quantum number J), i.e., one should have a two-level scheme with a Γ_8 quartet as the ground state and a Γ_7 doublet as the excited state,^{8,9} a picture which should be true as long as the environment of the trivalent Sm ions has cubic symmetry. The susceptibility data^{1,2} seem to be in line with this level scheme, the corresponding splitting energy ΔE being in the range $\Delta E \simeq 20$ K. But the specific-heat data¹ show apparently a more complex behavior with possibly more than just two levels and splitting energies in the range $\Delta E \simeq 25$ to 400 K: it can be interpreted perhaps in terms of a mixture of Sm^{3+} and Sm^{2+} ions.

Deuteron NMR measurements³ show an anomalous increase of the linewidth for $x > 0$ and $T < 50$ K which is interpreted in terms of magnetic fluctuations; such data are somewhat surprising because they occur at temperatures which are five times larger than the Néel temperature $T_N \simeq 10$ K. The neutron-diffraction and susceptibility data indicate also that the magnetic ordering may have an incommensurate component as soon as $x > 0$.^{3,5} All these properties suggest that the excess hydrogen concentration x modifies in an important way both the electronic and atomic structure of the dihydride. For not too large x values ($x \leq 0.25$), one expects *a priori* two main effects: first, a decrease of the conduction-carrier density, because each H atom tends to depopulate the conduction

band through the formation of a low-energy metal-hydrogen bond, a process which may also modify the shape of the Fermi surface and the corresponding nesting possibilities.¹⁰ Secondly, the crystal-field (CF) splitting may be modified, and this for two reasons: the excess H atoms may occupy the available octahedral interstitial sites in a statistical way, which can perturb the strictly cubic environment symmetry necessary for the occurrence of a two-level scheme; but there are also experimental observations, in TbH_{2+x} (Ref. 11) and PrH_{2+x} ,¹² that the excess H atoms may form an ordered structure (on a short-range or long-range scale). There are even indications, for CeH_{2+x} ,¹³ that this ordering may induce a cubic-tetragonal transformation of the fcc lattice of the rare-earth ions. Such a lattice deformation has apparently been observed under certain conditions for SmH_{2+x} when $x \geq 0.33$;⁶ NMR data indicate for $x > 0.3$ the occurrence of two lines below 240 K, which raises the question of whether it is due to the ordering process or to the presence of two phases.⁷ All these structural transformations may have of course a drastic influence on the CF splitting, i.e., they may for instance split the ground-state quartet into doublets.

In the present work we report on the electrical resistivity of SmH_{2+x} in the ranges $0 \leq x \leq 0.26$ and $1.5 \leq T \leq 300$ K. The resistivity $\rho(T)$ comprises a magnetic spin-disorder term ρ_m due to the exchange interaction $-\Gamma sS$ between the conduction-electron spins s and the localized rare-earth spins S . ρ_m is modified by the magnetic ordering for $T < T_N$, but also by the CFE for $T > T_N$: for the two-level scheme one expects the following behavior if Γ_8 is the ground state:

$$\rho_m = \begin{cases} 0.619\rho_m^0, & T_N < T \ll \Delta E \\ \rho_m^0, & T \gg \Delta E \end{cases}$$

(ΔE is the splitting energy between the Γ_8 and Γ_7 states, ρ_m^0 is the total spin-disorder resistivity¹⁴).

Of course, the CF splitting effect on $\rho_m(T)$ interferes

with the phonon and the eventual order-disorder contribution to the global resistivity $\rho(T)$. A good knowledge of the phonon contribution is necessary in order to separate the spin-order contribution. This has been achieved for the heavy rare earths^{15,16} but not yet for the light ones.

We shall also give some preliminary data concerning x-ray dilatation measurements as a function of x and of T .

EXPERIMENTAL

The electrical measurements were done on foils of size $20 \times 1 \times 0.2 \text{ mm}^3$, with four spot-welded platinum leads fixed on the metal before hydrogen loading. The hydrogen atoms are introduced in two steps: at about 500°C , we first prepare the dihydride whose exact composition $\text{SmH}_{1.98 \pm 0.01}$ is slightly substoichiometric; the additional hydrogen concentration x is absorbed at lower temperatures ($T \leq 300^\circ\text{C}$) under a pressure of the order of 10 Torr. The hydrogen concentration has been determined by measuring the pressure in a calibrated volume of the absorption system. The samples are then measured inside a pumped He cryostat by means of a classical dc method.

For the x-ray diffraction, the samples are in powder form and are maintained in a variable-temperature liquid-nitrogen cryostat.

RESULTS

We have followed the temperature dependence of a dozen $\text{SmH}_{1.99+x}$ specimens, with x varying between 0 and 0.26. Figure 1 gives a global view of the $\rho(T)$ variation for several selected values of x . For $x=0$, we ob-

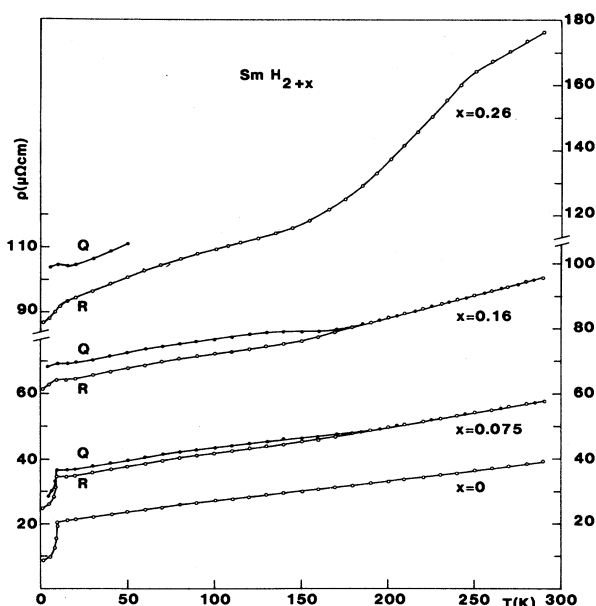


FIG. 1. Variation of the resistivity $\rho(T)$ of SmH_{2+x} specimens in the relaxed (R) and the quenched (Q) state over the whole temperature range.

serve a sharp magnetic transition at $T_N = 9.6 \text{ K}$ (see also Fig. 3) and apparently a classical variation of $\rho(T)$ for $T > 10 \text{ K}$ due essentially to the phonon contribution; there is no visible CFE contribution either in the low T range ($T < 40 \text{ K}$, Fig. 3) or in the higher T range ($T > 100 \text{ K}$), while such contributions are observed in TmH_2 (Ref. 15) and DyH_2 .¹⁶ It has to be remarked nevertheless that there is a positive curvature in the $\rho(T)$ variation of all heavy rare-earth dihydrides for $T > 200 \text{ K}$ which is due to the contribution of the optical phonons related to the vibrations of the H atoms.^{17,18} In the case of SmH_2 , we could expect an even greater curvature because of the higher lying multiplet J states (the corresponding excitation energy is of the same order as for the optical phonons, i.e., 1500 K).¹⁹ The absence of these terms may suggest that the optical phonons are at a somewhat higher frequency for the light rare earths; but it can also imply a CFE whose saturation on the $T \geq 200 \text{ K}$ range could suppress the expected positive curvature due to optical phonons.

For $x > 0$ (Fig. 1), it appears that $\rho(T)$ with increasing temperature is sensitive to the manner by which one reaches the low- T region: the resistivity is higher in the quenched state Q (cooling rate of about 10^3 K/min) compared with the relaxed state R (cooling rate of 0.3 K/min); the effect anneals at $T \leq 180 \text{ K}$. This indicates that an ordering process occurs within the octahedral sublattice similar to that observed for TbH_{2+x} ,^{11,12} but the corresponding amplitude of the difference between the Q and the R states is somewhat smaller in the case of SmH_{2+x} , especially for the $x = 0.26$ sample.

For $x = 0.075$ (Fig. 1), we observe again a sharp magnetic transition at the same temperature as for $x = 0$, for both the Q and the R state. For $x = 0.16$, the magnetic transition is somewhat less sharp and there is now a qualitatively different behavior for the Q and the R state (see also Fig. 4). Above $T \approx 180 \text{ K}$, one observes an increased slope compared to the case $x = 0$: it can be due either to a decrease of the conduction-electron density or to the order-disorder transition, which continues perhaps above 180 K in a way which is not dependent on the Q or R situation because of the higher diffusion rate at these temperatures.

For the $x = 0.26$ sample, we shall see (Fig. 5) that there is no clear evidence for a magnetic transition despite a rather sharp drop of $\rho(T)$ below $\approx 15 \text{ K}$. The most interesting feature for this composition concerns the very strong increase of $\rho(T)$ for $T > 160 \text{ K}$, which is certainly related to an order-disorder transition within the H sublattice (unfortunately, the voltage contacts had broken on the quenched sample above $T = 50 \text{ K}$). Nevertheless, it is to be noted that the residual resistivity ρ_r remains large for the R state, which was not the case for the $\text{PrH}_{2.25}$ compound:¹² the complete ordering is probably blocked by an insufficient diffusion rate below 160 K .

We have analyzed the annealing kinetics of the quenched state and show in Fig. 2, the behavior for $x = 0.075$ and 0.16 . There, we plot the difference $\Delta\rho_q$ between the Q and the R state; it shows a well-defined drop starting at about 130 K . It is interesting to note that the characteristic temperature at which the drop occurs is

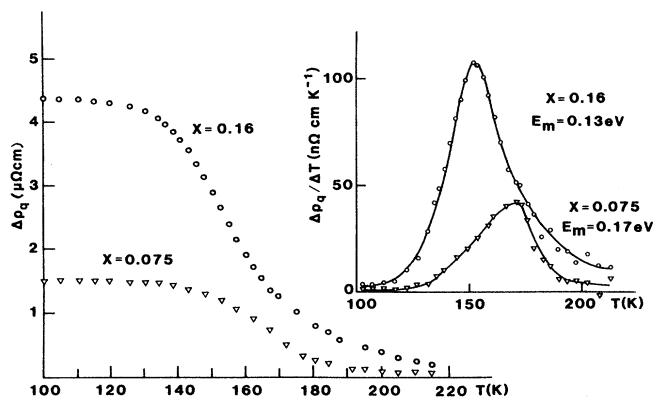


FIG. 2. Variation of the quenched-in resistivity $\Delta\rho_q$ as a function of temperature for two concentrations $x=0.075$ and 0.16 . The inset shows the derivatives $-d\Delta\rho_q/dT$ used for the determination of the activation energy E_m (see text).

clearly x dependent, namely it decreases with increasing x ; this is shown in the inset of Fig. 2 where we plot $d\Delta\rho_q/dT$ as a function of T ; the corresponding maximum occurs at 170 K for $x=0.075$ and at 153 K for $x=0.16$. By analyzing this recovery process (for details see Ref. 20) one can estimate the activation energy E_m for the migration of the recovering species; one finds $E_m=0.17(1)$ eV for $x=0.075$ and $E=0.13(1)$ eV for $x=0.16$. These data are in agreement with our earlier obtained results on PrH_{2+x} (Ref. 12) and with the NMR

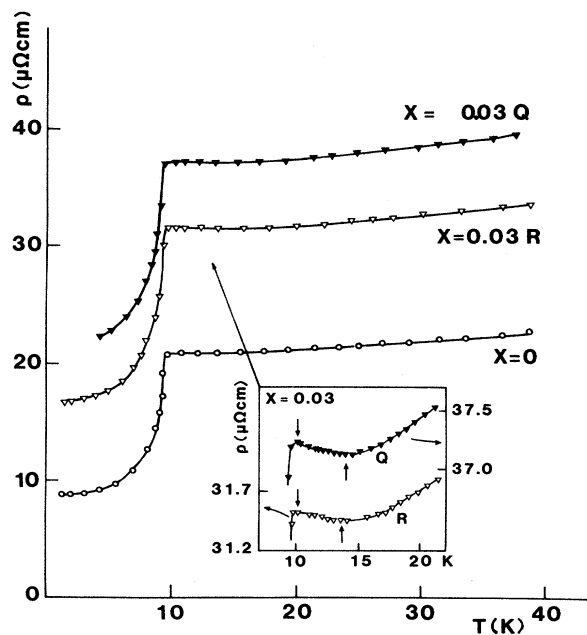


FIG. 3. Low-temperature resistivity variation for the quenched (solid signs) and relaxed (open signs) states for $x=0$ (no change in the Q state) and $x=0.03$. The inset shows the occurrence of a resistivity minimum for $x=0.03$, the arrows indicating the respective positions of the maxima and minima.

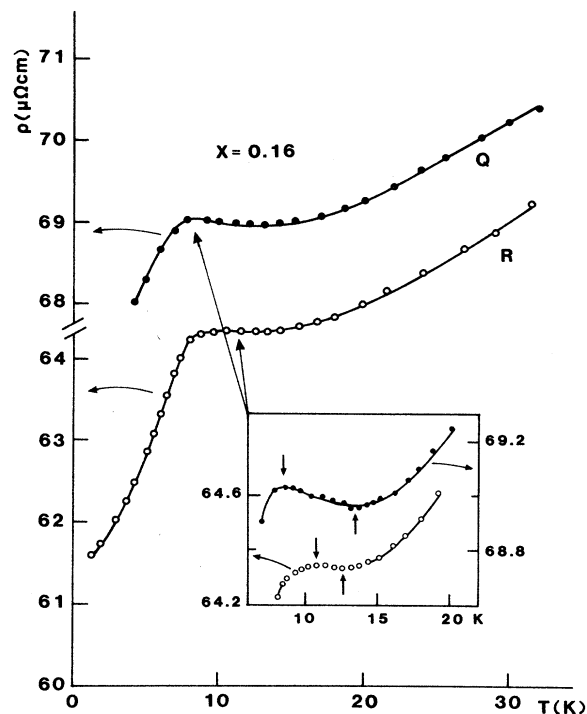


FIG. 4. Low-temperature resistivity variation for $x=0.16$ in the quenched (\bullet) and in the relaxed (\circ) state. The inset shows the increasing importance of the minimum after a quench.

data on LaH_{2+x} ,²¹ both showing a decreasing E_m with increasing x .

In Figs. 3, 4, and 5, we give some more detailed pictures of the $\rho(T)$ dependence in the $T \leq 40$ K range. The main observations are the following. First of all, as soon as $x > 0$, there appears a slight minimum in the $\rho(T)$ vari-

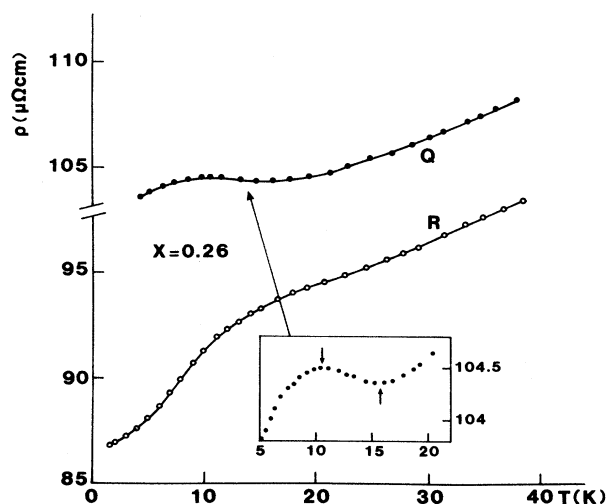


FIG. 5. Low-temperature resistivity variation for $x=0.26$ in the quenched (\bullet) and in the relaxed (\circ) state. The inset shows the appearance of a minimum in the quenched state.

ation situated between 12 and 16 K; its amplitude is always somewhat larger in the quenched state, specially for the $x=0.16$ and 0.26 samples. As concerns the absolute value of T_N , it does not change significantly with x for $x \leq 0.075$ from $T_N=9.6$ K; for $x=0.16$, one observes (Fig. 4) a transition onset at $T \simeq 10$ K but the true transition is likely to be near $T_N \simeq 8$ K. For $x=0.26$ (Fig. 5), it is not possible to define a clear transition temperature; there is a strong decrease of $\rho(T)$ for $T < 14$ K, but it does not have the shape of a well-defined transition, it decreases rather continuously down to 1.5 K (for the relaxed sample). This ρ diminution cannot be explained by the phonon contribution, which means that it is more likely related to CFE. The intensity of the magnetic interaction can also be tested by the values of the spin-disorder resistivity $\rho_m \simeq \rho(T_N) - \rho(0)$; we shall see that this value depends strongly on x (Fig. 6), in a way which is not coherent with the variation of T_N .

We have also done some preliminary x-ray determinations of the lattice parameter as a function of x and of T . First of all, we observe no change of structure [such as the cubic-tetragonal transformation observed for CeH_{2+x} (Ref. 13)] and no precipitation of the hexagonal high- x γ phase, which should occur for $x \gtrsim 0.5$,⁷ for the whole studied range up to $x=0.26$. There is nevertheless a qualitative difference between the behavior of the $x=0.26$ sample and that of the samples with smaller x , which concerns essentially the relative thermal expansion coefficient $(1/a)\Delta a/\Delta T = \alpha$ as a function of T . For $77 \text{ K} < T < 300 \text{ K}$, we observe an $\alpha(T) \simeq 3 \times 10^{-6} \text{ K}^{-1}$ for $x=0$ and $\alpha(T) \simeq 4 \times 10^{-6} \text{ K}^{-1}$ for $x=0.16$ in the whole temperature range; but for $x=0.26$ it appears that $\alpha(T)$ varies strongly; it is equal to 3×10^{-6} for $T < 150 \text{ K}$ and for $T > 250 \text{ K}$, while it takes an anomalous value $\alpha(T) \geq 10 \times 10^{-6} \text{ K}^{-1}$ in the range $150 \leq T \leq 250 \text{ K}$. We think that this variation is related to the order-disorder transformation within the octahedral H sublattice. A de-

tailed investigation of the x-ray diffraction including higher x values is on the way.

DISCUSSION

Absolute values of ρ_m and T_N

It is interesting to compare the values observed for SmH_{2+x} with those of GdH_2 , the dihydride for which one expects no CFE because the Gd ion has a pure spin moment. The absolute values of ρ_m and T_N are, respectively, $30 \mu\Omega \text{ cm}$ and 21 K in the case of GdH_2 .¹⁶ If one admits that the exchange integral Γ and the parameters of the electronic structure (the effective mass m^* and the carrier density n) are the same for all rare-earth dihydrides, a hypothesis which is relatively well tested for the heavy rare earths, then one expects that $\rho_m^0 \simeq (g_J - 1)^2 J(J+1)$ is equal to $8.5 \mu\Omega \text{ cm}$ (if one neglects the Γ_8 - Γ_7 splitting) or equal to $8.5 \times 0.619 = 5.26 \mu\Omega \text{ cm}$ if Γ_8 is the CF ground state. The observed value for $x=0$ is in fact much larger, namely $\rho_m = 12 \mu\Omega \text{ cm}$ (Fig. 6). Such a discrepancy relative to the de Gennes law has also been observed for PrH_2 :²² it indicates that Γ is perhaps larger for the light rare earths than for the heavy ones. It suggests also that the CF splitting is probably small for $x=0$, with $\Delta E \leq kT_N$, though we know that there is a curvature problem for the variation of $\rho(T)$ at high temperatures.

As concerns T_N , the expected value for SmH_2 is equal to 6 K while the observed one is 9.6 K , i.e., it can also be explained by a similar increase of Γ compared to GdH_2 .

More difficult to explain are the variations of ρ_m and T_N with x (or with the residual resistivity ρ_r , which depends on the sample state, R or Q). Figure 6 indicates that ρ_m increases first up to $\simeq 15 \mu\Omega \text{ cm}$ (corresponding to $x=0.05$ or $\rho_r = 22 \mu\Omega \text{ cm}$) beyond which it begins to

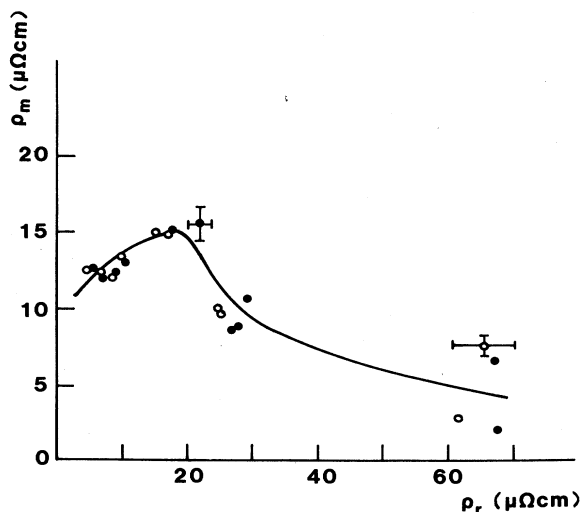


FIG. 6. Variation of the magnetic spin-disorder term $\rho_m = \rho(T_N) - \rho(0)$ as a function of the residual resistivity ρ_r , for relaxed (\circ) and quenched (\bullet) specimens.

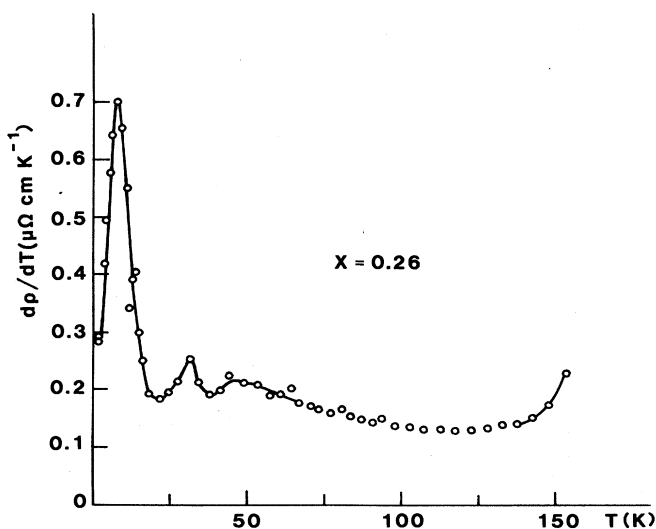


FIG. 7. Temperature variation of the slope $d\rho/dT$ for the relaxed $x=0.26$ sample. Note its decreasing tendency in the range 50 to 120 K, incompatible with a phonon contribution.

decrease quickly down to $\rho_m \simeq 3 \mu\Omega \text{ cm}$ for $x = 0.16$ (or $\rho_r = 61 \mu\Omega \text{ cm}$). But this is not really the case for T_N , which decreases only slowly from 9.6 K for $x = 0$ to $\simeq 8$ K for $x = 0.16$, while $T_N \simeq 0$ for $x = 0.26$. Of course, there are different parameters involved in the variation of ρ_m and T_N with x . The variation of $\rho_m^0 = (m^*/ne^2)\tau_s^{-1}$ involves band structure parameters (m^* and n) and the interaction relaxation time $(\hbar/\tau_s) \simeq (\Gamma^2/\epsilon_F)$. One must note also that if the magnetic ordering has an incommensurate component, then it may lead to an increase of ρ_r due to the opening of a superzone boundary gap, i.e., the measured value of $\rho_m = \rho(T_N) - \rho_r$ may have a diminished effective value; we make this remark because of the small minimum observed for $\rho(T)$. The value of T_N is also proportional to Γ^2/ϵ_F , but it involves furthermore the long-range Rudermann-Kittel interaction function $F(k_F r_{ij})$, where k_F is the Fermi wave vector and r_{ij} the distance between localized rare-earth spins; the variation of $k_F(x)$ may optimize this function and lead to a smaller decrease of T_N compared to that of $\rho_m(x)$. The proximity of k_F with a nesting condition¹⁰ could also affect T_N in a way which is very different from ρ_m .

Crystal-field effects

These effects are apparently very weak or even negligible for $x \simeq 0$ but are certainly important for $x = 0.26$. The absence of a real T_N can be due to a stronger splitting of the Γ_8 - Γ_7 levels or even—if the crystal field has a noncubic character—to a splitting of the Γ_8 quartet into doublets; all the possible corresponding ground states are always magnetic but with smaller magnetic moments. This latter qualitative idea is supported by the $\rho(T)$ variation up to 120 K (Figs. 5 and 7); anomalies of $d\rho/dT$ are observed near $T = 7.5$ K, 30 K, and even above 30 K. Without CFE, one would expect $d\rho/dT$ to increase (because of the phonon contribution) in this T range, while the observed decrease of $d\rho/dT$ can be related to a saturation of the CFE if condition $kT \geq \Delta E$ is satisfied, where ΔE is the splitting energy between CF levels.

Occurrence of a resistivity minimum

Well-defined resistivity minima attributed to incommensurate magnetic phases have been observed in TbH_{2+x} (Ref. 23) and PrH_{2+x} ,²² with amplitudes $\rho_{ic} = \rho(\text{max}) - \rho(\text{min})$ much larger than in the case of SmH_{2+x} : $\rho_{ic} \simeq 5-7 \mu\Omega \text{ cm}$ for TbH_{2+x} , $\rho_{ic} \simeq 0.7 \mu\Omega \text{ cm}$ for PrH_2 , $\rho_{ic} \leq 0.15 \mu\Omega \text{ cm}$ for SmH_{2+x} (Fig. 8). First of all, the proposed explanation for PrH_2 , where the minimum is probably due to CFE because of a magnetic ground state and a nearby nonmagnetic first excited state, is not applicable to SmH_{2+x} for two reasons: (i) all possible CF states in the case of SmH_2 are always magnetic states,⁹ and (ii) for PrH_{2+x} the minimum disappears with increasing x , while for SmH_{2+x} it is just the contrary, namely the amplitude of ρ_{ic} increases with x (Fig. 8). Qualitatively, the minimum effect resembles more to that of TbH_{2+x} where the ρ_{ic} amplitude increases also with x ; the fact that ρ_{ic} is always larger in the more disordered Q

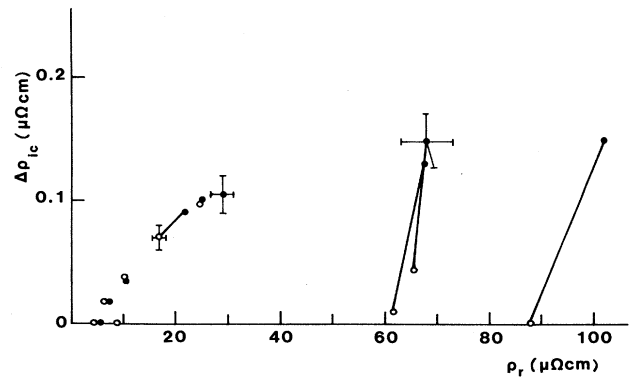


FIG. 8. Variation of the amplitude $\rho_{ic} = \rho(\text{max}) - \rho(\text{min})$ of the incommensurate contribution as a function of the residual resistivity ρ_r , for the relaxed (○) and quenched (●) states. The lines connect two states of the same sample.

state (particularly striking for the higher- x samples) agrees also with the observations on TbH_{2+x} . There has been noted a well defined antiferromagnetic but incommensurate ordering in TbH_{2+x} which could also be the case for SmH_{2+x} , though there are only indirect proofs for it.^{3,5} An important fact is the occurrence of the minimum even in the $x = 0.26$ sample (when quenched from room temperature) for which there is no real magnetic transition: this may indicate that the minimum is more likely related to incommensurate fluctuations of the magnetization M . It can be shown that such fluctuations are probably more important in the hydrides than in the pure rare-earth metals because of a modification of the parameter $k_F d$, where d is the nearest-neighbor distance between rare earth atoms. Critical fluctuations^{24,25} are only effective if $k_F d < \pi$, which is more likely to be the case for the hydrides because of the much lower carrier density in the dihydrides (one conduction electron per atom) compared to the pure metals (three conduction electrons per atom). The approach with x of an incommensurate nesting condition may of course also increase the amplitude of such fluctuations.

Concluding, we observe in SmH_{2+x} , for $x < 0.16$, a well defined magnetic transition with a large spin-disorder resistivity; this transition disappears for $x = 0.26$, which is probably due to a supplementary CFE (related to the octahedral H ions) capable to split the initial Γ_8 ground state. One of the most intriguing additional aspects concerns the observation of a resistivity minimum whose occurrence is probably related to the shape of the Fermi surface, the geometry of which reveals several possible nesting vectors;¹⁰ our data indicate that the approach towards a nesting condition is sensitive to the H concentration x (which modifies the electronic density) but also to the local atomic order or disorder of the H atoms, a sign that this also influences the electronic structure.

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