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Modulated magnetic properties in synthetic rare-earth Gd-Y superlattices

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Modulations of the magnetic properties in synthetic rare-earth Gd-Y superlattices were observed by magnetization measurements. The remanence and the saturation field show an oscillatory dependence on the intervening nonmagnetic Y layer thickness, but not on the ferromagnetic Gd layer thickness. The oscillation occurs with a periodicity of 7 atomic layers and extends over a range of 20 atomic layers. The results are interpreted on the basis of coherent propagation of the Ruderman-Kittel-Kasuya-Yosida interaction through the Y medium.

The synthesis of artificially layered materials has attracted much attention in the last decade. Studies of novel heterostructures tailored on length scales characteristic of important physical interactions have led to the discovery of many new physical properties.[1] In the case of magnetic superlattices, most studies reported to date have focused on the magnetic behavior at the interfaces between individual layers,[2,3] rather than on the long-range and collective phenomena representative of the superlattice as a whole.[4,5] This is largely due to the fact that many of the known magnetic interactions are of short range, and do not extend beyond several atomic planes. Earlier work on $3d$ magnetic superlattices such as the Cu-Ni systems found no evidence for a modulation effect on their magnetic properties.[6]

In comparison, the magnetic coupling in $4f$ magnetic rare-earth metals is relatively long range, and is mediated indirectly through the conduction electrons in the so-called Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Hence, magnetic superlattices containing magnetic rare-earth elements, e.g., Gd or Dy, alternating with a nonmagnetic analog such as Y, would seem to be promising systems for investigating the modulation effects derived from a long-range interlayer exchange coupling. Furthermore, previous studies have shown that rare-earth Gd-Y superlattices can be fabricated with a high degree of structural perfection and with well-characterized chemical and magnetic structures at the interfaces.[3,7] Therefore, the modulation effects can be readily explored by simply varying the intervening nonmagnetic layer thickness while maintaining a constant magnetic-layer thickness.

In this Rapid Communication, we report the first observation of significantly modulated magnetic properties in synthetic metallic superlattices. This is demonstrated by magnetization measurements on several series of Gd-Y superlattices where the remanence and the saturation field show an oscillatory dependence on the Y layer thickness with a periodicity of 7 atomic layers over a range of 20 atomic layers. The results are interpreted on the basis of coherent propagation of magnetic correlations across the nonmagnetic Y medium via the RKKY interaction which is enhanced by a peaklike structure in the Y generalized susceptibility function. Furthermore, the product of the wave vector at the maximum of this peak and the intervening Y layer thickness determines the sign of the magnetic interaction between adjacent Gd arrays and, therefore, the resulting magnetic structure of the superlattice.

$(\mathrm{Gd}_{N_{\mathrm{Gd}}}-\mathrm{Y}_{N_{\mathrm{Y}}})_M$ superlattices consisting of M successive bilayers of N_{Gd} atomic basal planes of hcp Gd followed by N_{Y} such planes of Y were grown by the molecular-beam-epitaxy techniques established for hcp rare-earth elements.[3] The magnetization studies were carried out at low temperatures by the Faraday method (1.4–12.8 kOe), and vibrating-sample magnetometry (0–15 kOe). The easy axis of the moment is in the plane, and the coercive field is typically less than 100 Oe. Previous studies focused on the magnetization behavior of the individual Gd array after reaching saturation.[3] The linear rise of the in-plane moment $\sigma_{\parallel}$ with the applied field was attributed to the nonferromagnetic interfacial Gd plane at either side of the ferromagnetic region.[7]

In this work two series of superlattices were examined: one consisting of $N_{\mathrm{Gd}}=10\pm1$ and $N_{\mathrm{Y}}=6-24$; the other of $N_{\mathrm{Gd}}=4$ and $N_{\mathrm{Y}}=2-10$. The results of the $N_{\mathrm{Gd}}=10\pm1$ series will be discussed first. At low fields, there appeared to be two different types of in-plane magnetization, shown in Fig. 1 for $T=12$ K. In the following discussions two characteristic parameters are introduced for describing the low-field magnetization behavior. The first parameter is the saturation field H_s at which $\sigma_{\parallel}$ no longer rises rapidly as it approaches saturation, and $\Delta\sigma_{\parallel}/\Delta H$ becomes constant. The other parameter is the ratio of $\sigma_r/\sigma(0)$, where σ_r is the remanence, and $\sigma(0)$ is the ferromagnetic moment at zero field, as determined by extrapolating $\sigma_{\parallel}$ from $H>H_s$ back to zero field.[3] The normalization of σ_r by $\sigma(0)$ removes the uncertainty in determining the total sample volume. In one case, represented by the $(N_{\mathrm{Gd}},N_{\mathrm{Y}})_M=(10,24)_{76}$ superlattice, the saturation is characteristic of ferromagnetic Gd thin films with weak magnetocrystalline anisotropy because of the zero angular momentum and the spin-only moment. The saturation field H_s is consistently low, about 2 kOe, and the ratio $\sigma_r/\sigma(0)$ is close to one. However, distinctly

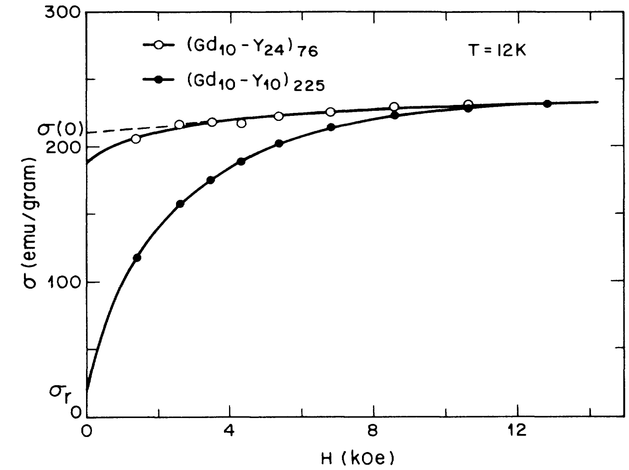

FIG. 1. The in-plane magnetization curves at 12 K for the $(10,10)_{225}$ and $(10,24)_{76}$ superlattices.

different behavior for the low-field magnetization is found in the $(10,10)_{225}$ superlattice: The remanent moment σ_r is exceedingly small, only about 10–15% of $\sigma(0)$. In addition, a much higher field of ~7 kOe is required to reach saturation.

Further studies of the in-plane magnetization behavior were carried out as a function of temperature. An equivalent method of presenting the data is to plot magnetic moment versus temperature in a series of applied fields ($H_{\parallel}$). Figure 2 shows the results for the (10,10) superlattice, where $H_{\parallel}$ is varied from 0.2 to 10.6 kOe. Instead of following the Brillouin function dependence of ferromagnetic Gd thin films, for $H_{\parallel}$ less than 7 kOe the moments develop a broad maximum at temperatures below the Curie point of 285 K. The occurrence of this moment maximum shifts to lower temperatures with increasing field, indicating a higher H_s at lower T. No evidence for the field-cooling effect is found. In light of recent neutron scattering studies,[5] the anomaly is now accounted for by the formation of a long-range magnetic antiphase domain structure. The spins of each Gd array lie predominantly in the plane. The increase of moment gained from the reduced thermal fluctuations at temperatures below T_c is compensated by an antiparallel arrangement of the spins of the adjacent ferromagnetic Gd arrays. Judging from the fact that H_s increases with decreasing temperature, this antiferromagnetic correlation becomes even stronger at lower temperatures.

The occurrence of the so-called "antiferromagnetic" behavior is not limited to the (10,10) samples, but is also found in the (11,9) and (10,16) samples in the same $N_{Gd}=10\pm1$ series, whereas the (10,6), (10,13), and (10,20) samples exhibit simple ferromagnetic behavior as in the (10,24) case. Studies were extended to another series of superlattices with $N_{Gd}=4$, and the data for moment versus temperature are shown in Fig. 3 for $N_Y=2$, 3, 4, and 10. The (4,10) sample has essentially the same magnetization behavior as the (10,10) sample except for a reduced $\sigma(0)$ value, which arises from the reduction of the density of the central ferromagnetic region in each Gd array.[3] In decreasing N_Y to 4 and 3, even a field exceeding 12.8 kOe is insufficient to preserve the Brillouin-like temperature dependence. Furthermore, the apparent maximum in M vs T shifts toward higher T with decreasing N_Y, accompanied by a substantial loss of moment at lower T. This suggests an antiferromagnetic order progressively more difficult overcome by the field. For $N_Y\leq 2$, the fact that the relatively modest values of σ_r and H_s reoccur in the (4,2) sample implies a reversed trend toward ferromagnetic coupling. Note that in these samples of small N_Y, interdiffusion near the interfaces may cause the chemical modulation amplitude to be significantly reduced below 100%. Nevertheless, present magnetization data are both qualitatively and quantita-

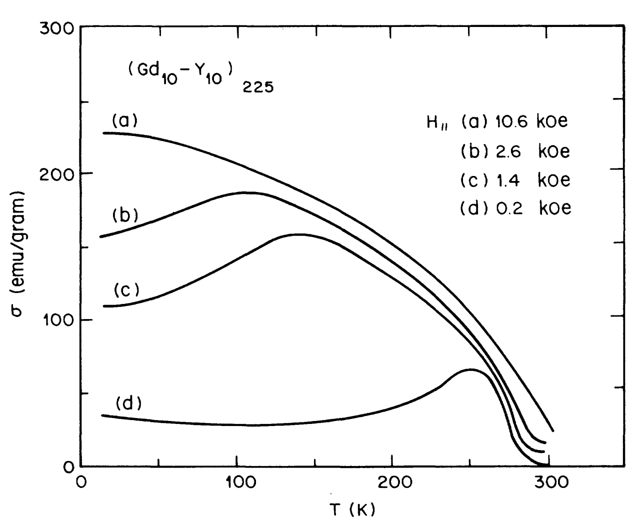

FIG. 2. The temperature dependence of the magnetic moment of the (10,10) superlattice in a series of applied fields.

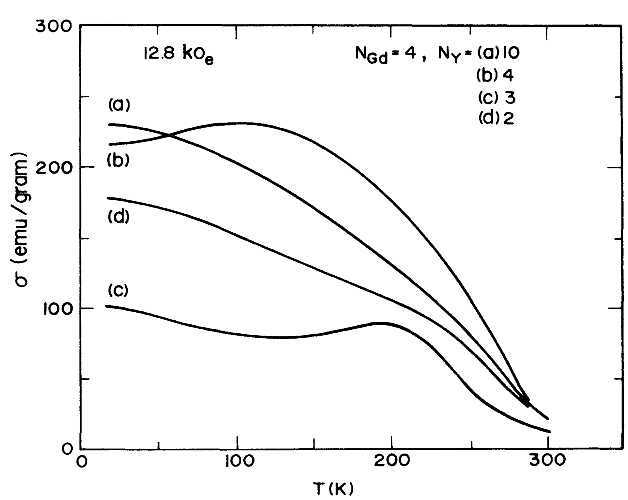

FIG. 3. The temperature dependence of the magnetic moment of the (4,2), (4,3), (4,4), and (4,10) superlattices in an applied field of 12.8 kOe.

tively different from those expected for Gd-Y alloys of the same average compositions,[8] or of compositionally modulated Gd-Y alloys, e.g., $Gd_{0.7}Y_{0.3}$ alternated with $Gd_{0.3}Y_{0.7}$.

The modulation effect on the magnetic properties is best demonstrated by the variations of $\sigma_r/\sigma(0)$ and H_s with N_Y, plotted in Figs. 4(a) and 4(b), respectively. The inverse relationship between $\sigma_r/\sigma(0)$ and H_s is evident. Most strikingly, the data for both parameters are consistent with a trend exhibiting an oscillatory dependence on N_Y for $2 \leq N_Y \leq 20$. The oscillation period is approximately 7 atomic layers, and the overall oscillation range is about 20 atomic layers. Furthermore, this oscillatory dependence of the magnetization on N_Y is not limited to two series of superlattices. The data shown in Fig. 4, in fact, represent a much larger body of results obtained on superlattices of different N_{Gd} including 3, 5, and 20. Varying N_{Gd}, to first order, does not affect the oscillatory trend qualitatively.

It is interesting to speculate on the nature of the fundamental mechanism governing the modulated magnetic behavior in the superlattices. The most appealing explanation that consistently accounts for the oscillatory dependence is based on the picture of a long-range RKKY interaction propagating coherently through the nonmagnetic Y medium which then causes an effective coupling between the successive Gd arrays. This postulation stems from the known properties of the diluted alloys of magnetic rare-earth elements with Y. With the addition of low concentrations of magnetic rare-earth ions to nonmagnetic Y, a long-range antiferromagnetic in-plane helical order is formed.[9] The turn angle of the helix remains the same regardless of the magnetic species added. In the very dilute limit (~2%), the turn angle of the spiral is 50.4° per atomic layer. The formation of this long-range magnetic order was attributed to RKKY coupling mediated by the conduction electrons of Y, with a wavelength determined by the peak in the generalized susceptibility $\chi_Y(q)$ of Y at $q_{max}=0.28(2\pi/c)$.[10]

Based on this picture, an interpretation of the present data is therefore made. The susceptibility function $\chi_{Gd\text{-}Y}(q)$ of the overall superlattice is approximately some average of the susceptibilities of the two components. The peaklike structure at q_{max} in $\chi_Y(q)$ is expected to be directly reflected in $\chi_{Gd\text{-}Y}(q)$ and, therefore, in the exchange function $J_{Gd\text{-}Y}(q)$, since $\chi_{Gd}(q)$ has a maximum at $q=0$. Consequently, the sign of the exchange interaction function $J_{Gd\text{-}Y}(r)$ will depend in an oscillatory way on the Y layer thickness with a periodicity of ~7 atomic layers corresponding to $2\pi/q_{max}$. An estimate for $J_{Gd\text{-}Y}(r)$ vs N_Y was made, and shown in Fig. 4(c).[5,11] The calculation is for a simplified case of two monatomic Gd planes separated by a Y layer of variable thickness. $\chi_{Gd}(q)$, originally responsible for propagating the indirect RKKY coupling in Gd, is now replaced by $\chi_Y(q)$. The calculated $J_{Gd\text{-}Y}(r)$ approximates that of the superlattices when only the nearest-neighbor exchange interactions among Gd arrays are considered.

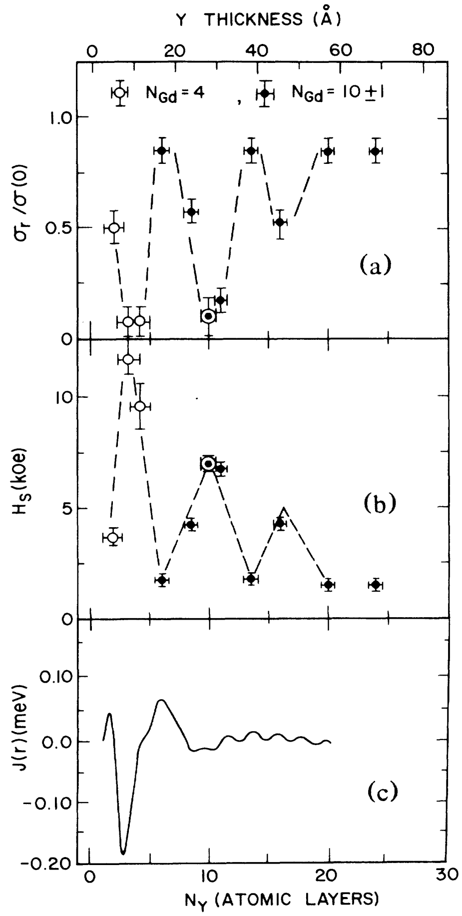

FIG. 4. The oscillatory dependence of (a) $\sigma_r/\sigma(0)$, and (b) H_s, on N_Y in two series of superlattices with $N_{Gd}=4$, and $N_{Gd}=10\pm1$. The dashed lines are to guide the eyes. (c) The calculated functional dependence of $J_{Gd\text{-}Y}(r)$ on N_Y.

Since the exchange fields of the ferromagnetically aligned Gd planes within each array are colinear in one direction, the presence of a negative exchange interaction between the adjacent Gd arrays produces an antiferromagnetic alignment (i.e., 180° apart) between these arrays. This antiferromagnetically ordered structure corresponds to a magnetization behavior of low remanence and high saturation field. The interpretation is in good agreement with the experimental observation of the periodic occurrence of the low $\sigma_r/\sigma(0)$ and high H_s at $N_Y=3$, 4, 10, 11, and 16. Note that a deviation from the expected square-wave modulation exists in $\sigma_r/\sigma(0)$ vs N_Y. In the (11,9) and (10,16) samples, $\sigma_r/\sigma(0)$ is about 0.5, and the antiphase domain spin structure was found in neutron diffraction. Two possible explanations are proposed to account for such behavior. The first possibility considers that $J_{Gd\text{-}Y}(r)$ in such N_Y range is of negative sign, but of weaker magnitude, according to Fig. 4(c). After magnetizations at high fields, the spins of successive Gd arrays may return, at zero field, to an arrangement of being canted symmetrically with respect to the field with an angle θ. Here θ could be approximately 60° to give a $\sigma_r/\sigma(0)$ ~0.5. The other possibility is based on the fact that $J_{Gd\text{-}Y}(r)$ changes sign rapidly near such N_Y range. Minor fluctuations in N_Y readily resulted in coexistence of ferromagnetic and antiferromagnetic alignments between adjacent Gd arrays in the overall sample, hence given an average remanence about 50% of $\sigma(0)$. Detailed analysis of the polarized return diffraction data would resolve these two different spin structures.

In addition, consistency was observed between the calculated dependence of $J_{\text{Gd-Y}}(r)$ on N_{Y} and the experimental data for H_s, since H_s can be taken as a measure of the exchange coupling strength between the Gd arrays. The overall decrease of H_s with increasing N_{Y} follows approximately $1/r$, again, consistent with the dependence of the RKKY coupling in the one-dimensional case. Moreover, because of the finite width of the peak in $\chi_{\text{Y}}(q)$, the interaction range of $J_{\text{Gd-Y}}(r)$ is limited to be about 20 atomic layers, which is also supported by the data in Figs. 4(a) and 4(b).

In conclusion, this work has demonstrated for the first time that the magnetic properties representative of synthetic superlattices as a whole are modulated by artificially imposed periodicities. Further, novel magnetic properties are expected to emerge continually, as studies can now be straightforwardly extended to many other systems by introducing different species of magnetic and nonmagnetic components, and even nonperiodic layering sequences.

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