Magnetic ordering of EuTe/PbTe multilayers determined by x-ray resonant diffraction

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In this work we use resonant x-ray diffraction combined with polarization analysis of the diffracted beam to study the magnetic ordering in EuTe/PbTe multilayers. The presence of satellites at the $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$ magnetic reflection of a 50 repetition EuTe/PbTe superlattice demonstrated the existence of magnetic correlations among the alternated EuTe layers. The behavior of the satellites intensity as T increases toward the Nèel temperature T_N indicates that these correlations persist nearly up to T_N and suggests the preferential decrease of the magnetic order parameter of external monolayers of each EuTe layer within the superlattice. © *2008 American Institute of Physics*.
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X-ray magnetic resonant diffraction is a useful technique for the study of magnetic materials.[1] In resonant diffraction, the natural weakness of the magnetic diffraction is partially overcome by the intensity enhancement of several orders common at L and M absorption edges of rare earths and transition metals.[2–4] Availability of synchrotron sources with bright, white, polarized, and collimated beams has been fundamental for the increasing applications of x-ray magnetic resonant diffraction. Compared to the well established neutron diffraction, x-ray magnetic diffraction has several advantages such as the higher resolution and surface sensitivity. Thin magnetic films and multilayers show interesting phenomena such as interlayer exchange coupling (IEC) across nonmagnetic spacer,[3,5,6] and surface magnetization effects.[7] In this work, we use resonant x-ray diffraction to study the magnetic order in EuTe/PbTe multilayer structures, demonstrating the capability of the technique to detect IEC and near surface magnetization profiles. EuTe is a wide gap semiconductor and a classical Heisenberg antiferromagnet (AFM) with Nèel temperature T_N=9.6 K.[8] The magnetic properties arise from the half filled Eu ion 4f electron shell, with $S=\frac{7}{2}$. The magnetic moments align parallel within (111) lattice planes, and antiparallel between adjacent planes. Hence, below T_N half order (hkl) magnetic peaks are expected to appear.

The samples were grown on (111)BaF_2 substrates by molecular beam epitaxy (MBE) in a Riber 32P MBE system. The magnetic x-ray diffraction measurements were made at the XRD2 line at the Brazilian Synchrotron facility.[9] The sample was mounted into a two cycle ARS cryostat able to reach 1.5 K, fixed itself on a six-circle Huber diffractometer with vertical diffraction plane. Beam flux at the energy used was $\sim 5\times10^{10}$ photons/s, focused on a 0.6 mm vertical $\times$2 mm horizontal spot at the sample position.

In order to tune the energy to the resonance expected near the Eu L_{II} absorption edge, the magnetic diffraction of a 1.5 μm EuTe film grown directly on a BaF_2 substrate was studied, using a Canberra solid state detector with 160 eV energy resolution. Below T_N, magnetic peaks appeared at half order $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$, $(1\frac{1}{2}\ 1\frac{1}{2}\ 1\frac{1}{2})$, etc., reciprocal lattice points (rlp). A resonant enhancement in the half order peaks intensity of more than two orders was found at 7611 eV, 2 eV above the Eu L_{II} absorption edge at 7609 eV, determined as the maximum of the fluorescence derivative. The half order Bragg peaks were tested by a polarization analysis with a graphite analyzer, showing that the peaks were purely $\sigma\rightarrow\pi$ scattered, consistent with their magnetic origin.

We started investigating a 40 monolayers (ML) EuTe film grown on a PbTe 3 μm buffer layer, and covered with a 50 ML PbTe cap. A polarimeter with a graphite analyzer and a Cyberstar scintillation detector was installed on the 2θ arm of the Huber diffractometer, allowing to rotate the analyzer diffraction plane around the 2θ arm. When the analyzer diffraction plane was perpendicular to the sample diffraction plane, and the graphite (003) reflection was chosen ($2\theta_{003}=93.3°$ at 7611 eV), $\sigma\rightarrow\pi$ magnetically scattered intensity from the sample was recorded. This configuration keeps charge $\sigma\rightarrow\sigma$ scattered intensity from reaching the detector, thus eliminating most of the background noise, which allows measuring the weak magnetic signals coming from the sample.

Figure 1(a) shows the scan across the (222) structural reflection measured in the $\sigma\rightarrow\sigma$ channel. The three peaks from right to left correspond to the BaF_2 substrate, the PbTe buffer layer, and the EuTe film. The fit to the measurement, using dynamical diffraction theory, yielded a thickness of 40$\pm$1 ML for the EuTe film. Scanning the $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$ rlp in the $\sigma\rightarrow\pi$ channel, a clear magnetic peak was observed [Fig. 1(b)]. This time a kinematical model was used to fit the measurement,[6] justified by the weakness of the magnetic diffraction. An almost perfect match was obtained considering a 40 ML AFM ordered EuTe layer.

After checking the suitability of the technique to study very thin EuTe films, we went on with a 50 repetitions EuTe/PbTe superlattice (SL), grown on a 3 μm PbTe buffer layer. Individual thicknesses of 7 and 14 ML ($\pm$0.5 ML) for EuTe and PbTe layers within the SL were found from the

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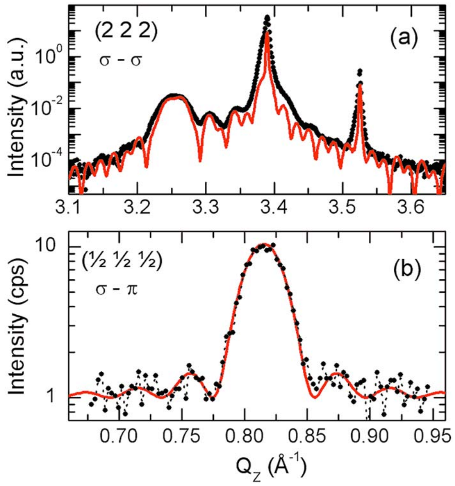

FIG. 1. (Color online) $\theta/2\theta$ scans along the (2 2 2) structural and $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ magnetic reflections of a 40 ML EuTe film (dots) measured at 1.5 K in the $\sigma\rightarrow\sigma$ and $\sigma\rightarrow\pi$ channels, respectively. The solid lines are the dynamical (a) and kinematical (b) fits to the measurements.

dynamical fit to the measurement across the (222) structural reflection, measured in the $\sigma\rightarrow\sigma$ channel [Fig. 2(a)]. For the fit, we considered in-plane strains of −0.017 and 0.004 for EuTe and PbTe layers, respectively, as obtained from the reciprocal space map around the (224) reflection. The SL satellite intensities decay faster than expected according to the simulation [Fig. 2(a)], which is ascribed to interface roughness not considered in the model.[10]

Changing to the $\sigma\rightarrow\pi$ channel, a well-resolved satellite structure was observed near the $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$ rlp [Fig. 2(b)]. The satellite structure proves the existence of magnetic correlations among the alternated EuTe layers. The kinematical model described in Ref. 6 was used to fit the measurement and obtain the type of magnetic correlations present. The fit yields a parameter p, whose sign indicates if the monolayer

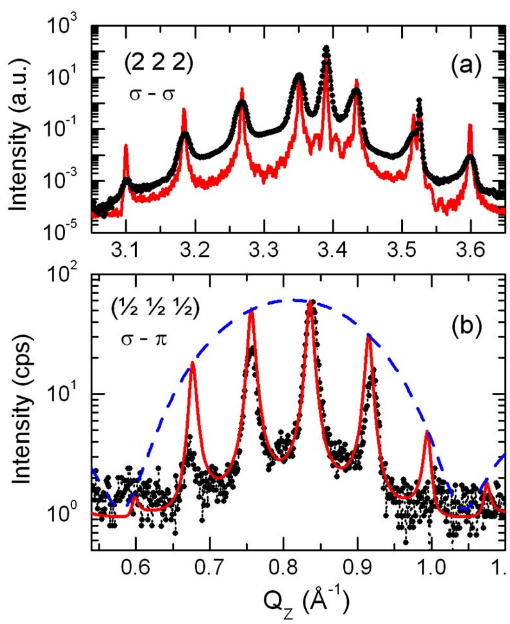

FIG. 2. (Color online) $\theta/2\theta$ scans along the (2 2 2) structural and $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ magnetic reflections of a 50× (7 ML EuTe/14ML PbTe) SL (dots) measured at 1.5 K in the $\sigma\rightarrow\sigma$ and $\sigma\rightarrow\pi$ channels, respectively. The solid lines are the dynamical (a) and kinematical (b) fits to the measurements. The dashed line (b) is the magnetic structure factor of a 7 ML EuTe layer.

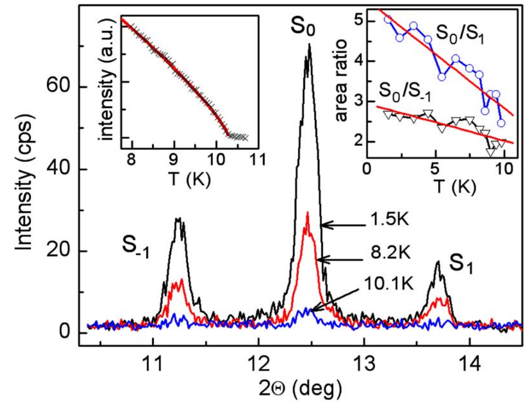

FIG. 3. (Color online) $\theta/2\theta$ scans along the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ magnetic reflection of a 50× (7 ML EuTe/14ML PbTe) SL, measured at 1.5, 8.2, and 10.1 K. The left inset shows the intensity at the central satellite position vs temperature (×), with a power law fit (solid line). The right inset shows the temperature dependence of the integrated intensity ratio of the central satellite (S_0) to the two outer satellites (S_1 and S_{-1}).

magnetization sequence in layer i is equal (in-phase configuration, $p>0$), or reversed (out-of-phase configuration, $p<0$) to that of layer $i+1$. The modulus of p equals the probability that the magnetic sequence is maintained by any two adjacent EuTe layers. Our fit yielded $p=-0.7$, indicating that the magnetic sequence in adjacent EuTe layers is reversed.

The magnetic satellites intensity lowered as the temperature increased toward T_N (Fig. 3). Fitting the temperature dependence of the intensity at the central satellite position with a power law $(T-T_N)^{2\beta}$ (left inset of Fig. 3) yielded $T_N=10.3$ K and a critical exponent $\beta=0.4$. The satellite structure persisted almost up to T_N (Fig. 3), indicating that the interlayer correlations continue to exist while intralayer magnetic ordering exists. The extent of the correlations also remains constant, since no widening of the satellites was observed. Hence, the possibility of an intermediate uncorrelated state just below T_N as suggested in Ref. 11 can be either discarded or reduced to a very small temperature interval.

The right inset of Fig. 3 shows that the integrated intensity ratios of the central satellite to the outer satellites decrease with temperature. This can be interpreted considering that the intensities are modulated by the structure factor of a 7 ML EuTe magnetic layer [dashed line in Fig. 2(b)]. If the magnetic order parameter profile was homogeneously reduced within each EuTe layer as the temperature increases, the modulating structure factor would only change by a constant. This would lead to weaker satellites, with constant relative intensities. The faster intensity decrease of the central satellite relative to the outer satellites means that the modulating structure factor is widening, which results from a narrowing of the magnetic profile, since they are related by a Fourier transform. We can thus infer that the magnetic order parameter decreases preferentially in the external monolayers of each EuTe layer as the temperature increases toward T_N.

To conclude, we demonstrated the suitability of magnetic resonant x-ray diffraction for the routine study of very thin EuTe films and SLs, and for the detection of magnetic correlations in AFM SLs, which had been done only by neutron diffraction.[6,12] The intensity gain obtained with resonant diffraction, together with the use of a polarimeter to virtually suppress any charge background, allowed measuring clear magnetic diffraction peaks, even at temperatures close to T_N, giving insight on the magnetic order parameter profile.

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