Spin-resolved electronic structure of EuO across the Curie Temperature

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The electronic structure of the ferromagnetic semiconductor EuO is investigated by means of spin- and angle-resolved photoemission spectroscopy and density functional theory. Valence bands of EuO exhibit unique properties of hosting both the fully exchange split Eu 4f, as well as O 2p levels. Our spin-resolved data directly demonstrates the exchange splitting in O 2p and reveals that, while the macroscopic magnetization of the sample vanishes at the Curie temperature \( T_C \), the experimentally-determined exchange splitting of the O 2p band persists up to \( T_C \); if the picture of fluctuating spin-blocks is assumed. We discuss possible temperature-related spectral changes by analyzing ferromagnetic and antiferromagnetic phases, directional effect due to spin-orbit coupling, as well as effects due to sample aging, unavoidable for this highly reactive material. Our calculations with a Hubbard \( U \) term reveal a complex nature of the local exchange splitting on the oxygen site and in conduction bands, shining a new light on the interpretation of previous optical and photoemission spectroscopic results.

I. INTRODUCTION

Nowadays, it is accepted [1] that the intra-atomic exchange splitting \( 2\Delta E_{ex} \) (where \( \Delta E_{ex} \) is the exchange energy) persists above the magnetic ordering temperature in most materials, laying the foundation for the framework of local magnetic moments (LMs). The existence of short-range order in itinerant magnets [2–5] is being described within this framework, as well as numerous important magnetic phenomena such as the nature of the magnetic order in dilute magnetic topological insulators [6, 7] or the surface ferromagnetism in novel 4f compounds [8, 9]. In the ferromagnetic (FM) phase, at a temperature far below the Curie temperature \( (T_C) \), LMs are all aligned parallel. A global magnetic phase collapses above the ordering temperature, and one of the fundamental problems of condensed matter physics concerns the existence and the nature of a short range magnetic order in the paramagnetic phase [1–5, 10–14]. Different, but largely complementary, pictures have been used to model the vanishing macroscopic spontaneous magnetization at \( T_C \). In particular, two limiting descriptions can be considered. One is the limit of fully disordered LMs [13], which in the ‘language of magnons’ is an equal occupation of all magnon modes. The second is the limit of fluctuating spin blocks [14], which is the limit where only low \( q \) magnons (long wavelength spin waves) have significant occupation. Advanced theoretical treatments of LMs based on the coherent potential approximation (CPA) [13, 15] are single-magnetic-site descriptions that routinely do not take into account the short range order [3, 10]. Nowadays, the evidence for short range magnetic order above \( T_C \) comes not only from the susceptibility studies [16], but also from methods such as the neutron diffraction [17], electron energy loss spectroscopy [18], and inverse photoemission [19].

In this work, we are studying the electronic structure of EuO across the ferromagnetic-paramagnetic phase transition. We have chosen EuO as a prototypical Heisenberg FM, offering a benefit of the easily accessible Curie temperature \( (T_C \sim 70\text{K}) \). On the other hand, EuO is a highly reactive material, posing significant experimental challenges, in spite of keeping the highest standards of the ultra-high vacuum methodology. To circumvent these problems, we have employed spin-resolved version of angle-resolved photoemission (spin-ARPES) to get access to the configuration-averaged electronic structure across \( T_C \).

Spin- and angle-resolved photoemission (spin-ARPES) has been the method of choice for studying the band structure of magnetic materials [20–22]. We are only aware of a single report on spin-resolved photoemission on EuO: in an angle-integrated study it was reported that the spectral polarization is temperature dependent and vanishes at \( T_C \) [23]. In the case of spin-integrated ARPES, a recent synchrotron study on Gd-doped EuO has reported for the first time results where clear dispersions in the O 2p manifold are present in experimental \( E(k) \) maps [24]. Earlier work by Miyazaki et al. [25] reported a vanishing of the \( 2\Delta E_{ex} \) in O 2p at \( T_C \), based on

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the analysis of the second derivative of ARPES spectra at \( \Gamma \) and X points of the Brillouin zone (BZ). Furthermore, the well-established red shift of the optical absorption edge [26, 27], the temperature-dependent resistivity of EuO tunnel junctions [28, 29], and the metal-insulator transition (MIT) in Eu-rich samples [30, 31] have been ascribed to the vanishing of the \( 2\Delta E_{ex} \) in the Eu 5d-derived conduction band minimum (CBM) at \( T_C \approx 70 \) K [32].

In order to get more insight into the theoretical aspects of the relation between the magnetic configuration and the electronic structure, we have employed the \textit{ab initio} calculations. Band structure calculations for FM, and two antiferromagnetic phases of EuO, AFM-I, and AFM-II [33, 34], visualize the band renormalizations under various magnetic arrangements of Eu 4f moments. The AFM arrangements can be considered as a limiting case, a hypothetical full occupation of magnons at the highest \( \mathbf{q} \). In addition, we analyze the role of spin-orbit coupling (SOC), which leads to a directional dependence of the band energies. We find this effect to be particularly important for the Eu 4f levels.

Based on the analysis of the temperature-dependent spectral renormalization in our spin-intergrated ARPES we propose a complex interplay of effects due to vanishing magnetization, SOC, phonon effects, and surface aging, preventing unambiguous interpretation of the standard ARPES data. To solve this problem, we employ spin-ARPES, which at \( T < T_C \) is able to disentangle magnetic and non-magnetic spectral contributions and probe the temperature-dependent spectral renormalization and \( 2\Delta E_{ex} \) in the occupied valence bands. The exchange splitting in O 2p is directly demonstrated, showing two exchange split doublets, as expected from the theoretical predictions. A conventional space- and time-averaging analysis suggests that at \( T_C \) the global magnetization and the exchange splitting in O 2p both vanish. An alternative analysis made under the assumption of fluctuating spin blocks reveals that the exchange splitting in O 2p does not follow the vanishing of the global magnetization and remains nearly constant up to \( 60 \) K.

Taking into account these findings, we propose that at \( T_C \) EuO remains in the regime of the fluctuating spin blocks with the spin-correlation length large enough to sustain local exchange splitting in the band structure. At \( T > T_C \) the possible persistence of the exchange splitting should depend on the spin-correlation length, that would enable forming the exchange-split band structure locally.

II. EUO BAND STRUCTURE IN FM, AFM-I, AND AFM-II PHASES

Realistic DFT+U band structures of EuO in FM and AFM phases are presented in Fig. 1. These calculations were performed using the full-potential linearized augmented plane wave method as implemented in the WIEN2k code [35] at the cubic lattice constant of 5.14 Å. For the exchange-correlation functional we used the generalized gradient approximation (GGA) [36]. In order to correctly describe the relative positions of bands as measured in photoemission experiments, we have included static local electronic correlations to the GGA potential in the GGA+U method [37] with \( U = 7.9 \) eV on Eu 4f orbitals.

The symbol size in Fig. 1 indicates the local O 2p character on a particular site, while blue (red) color mark the predominantly majority (minority) spin character. The calculated cases are: the FM phase with all Eu spins aligned, the AFM-I with 4 Eu nearest neighbours (NN) and 2 NN in opposite spin directions, and the AFM-II where 3 NN Eu are spin-up and the other 3 spin-down [34]. AFM-I, and AFM-II phases can be seen as spin spirals at highest \( \mathbf{q} \) along the [001] direction, and highest \( \mathbf{q} \) along the [111] direction, respectively. The insets in Fig. 1 show schematics of these magnetic configurations with bigger blue (red) balls as Eu majority (minority) ions, and small balls as oxygen ions. In addition, the Supplementary Fig. S1 visualizes the influence of the FM and AFM-I phases on the spectral shapes and bandwidths by directly comparing the bands along the same reciprocal directions, as referred to the crystal lattice, for the two phases. Detailed plots of various orbital characters and additional comparisons are provided in the Supplementary Material (Fig. S2), and the picture that emerges reveals a complex character of energy splittings in the O 2p manifold: the exchange splitting depends not only on the crystalline direction but also on the orbital character of the electronic states.

Importantly, in the AFM-I phase, locally, the O 2p orbitals are partially polarized, and when looking at a particular O site in the lattice, the polarization order reverses depending on the orbital character. Without the loss of generality, let us assume that the intralayer FM arrangement is within \( x-y \) planes, see a schematic crystal lattice next to Fig. 1(c). Along \( z \), subsequent layers are arranged antiparallel. Therefore, despite the fact that O 2p are locally polarized their global polarization of course vanishes. Then, O 2px and 2py interact with Eu 4f moments within the same plane, and O 2pz interacts with Eu 4f moments from adjacent planes. In the FM case the real \( p_x, p_y \) and \( p_z \) orbital character is mixed for all bands with the O 2p dispersion reminiscent of the valence bands of the classical semiconductors [38], but for AFM-I along many high-symmetry directions O 2p bands split according to real orbitals in the cubic basis (suggesting \( \pi- \) and \( \sigma- \) like bonding for AFM phases). This leads to the spin reversal in bands derived from in-plane and out-of-plane (in our geometry) O 2p orbitals. As an example we have highlighted bands along \( \Gamma-X \) by dashed line boxes in Fig. 1(c) and (d). The order of red-blue colors reverses between these two cases, in particular near the \( \Gamma \) point, which means their spin polarizations are anti-parallel, with the local polarization reaching nearly 100%.

The understanding of the magnetism in Eu chalco-
FIG. 1. DFT+$U$ band structure and the corresponding BZs of (a) FM, (b) AFM-II and (c)-(d) AFM-I phases of EuO. Energy eigenvalues are depicted with green lines. The size of colored filled circles indicate the $p$ orbital character of the oxygen atom, and colors depict the spin-polarization in these orbital projections. Lattice spin up/dn configurations of AFM-II and AFM-I phases are shown next to panels (b) and (c) respectively. The two regions marked by dashed line boxes in (c) and (d) indicate a reversal of the spin character for differently dispersing bands.

Spin-ARPES experiment is always performed on the ensemble of the photoemitted electrons, and is able to determine a spin component along the quantization axis of the analyzer. The measurement influences the spin state through the wave function collapse, therefore it is not possible to experimentally distinguish e.g. between the ensemble of randomly oriented spins and the ensemble of half-half distributed antiparallel spins, as both cases will

The analysis of the band characters in Fig. 1 (and Supplemental Fig. S2) can provide additional insights into the hybridization between particular band manifolds both in FM and AFM phases. As shown for the AFM-I phase in Fig. 1(c)-(d), the exchange splitting of e.g. O $2p_z$ orbitals depends on the spin orientation of the Eu $4f$ spins from adjacent layers. The case for Eu $5d$ is similar, with the difference in the $5d$ orbital shapes that despite being centered on Eu sites can overlap with both their own on-site $4f$ and some of their 12 NN $4f$, depending on the $d$ orbital symmetry. The non-vanishing local spin polarization in the Eu $5d$ also for the AFM-II phase (Supplemental Fig. S2) seems to be related to this overlap. Therefore, the temperature dependence of the exchange splitting in O $2p$ bands is expected to be somewhat different than in Eu $5d$.

III. BAND RENORMALIZATIONS DUE TO SPIN-ORBIT COUPLING

Spin-ARPES experiment is always performed on the ensemble of the photoemitted electrons, and is able to determine a spin component along the quantization axis of the analyzer. The measurement influences the spin state through the wave function collapse, therefore it is not possible to experimentally distinguish e.g. between the ensemble of randomly oriented spins and the ensemble of half-half distributed antiparallel spins, as both cases will
FIG. 2. Theoretical DFT calculations with SOC included. (a) The band structure along the [001] Γ − X direction (in our case the direction normal to the surface), for the magnetization $M$ along [111] (solid line) and [001] (dashed line). (b) Spin polarized partial density of states (pDOS) for the [001] Γ − X direction for $M$ along [111], red and blue lines depict minority and majority character, black line shows the spin integrated pDOS. (c)-(d) pDOS along the [001] direction for the O 2p and Eu 4f manifolds respectively with 20 meV Gaussian broadening. The black line shows the pDOS for $M$ along [111], and the magenta line shows the normalized sum of the pDOS for $M$ along all the <001>, <110> and <111> directions in order to approximate blocks of random $M$ orientation, while performing the normal emission ARPES.

yield the same result [45]. Therefore, if only the isotropic exchange is considered (and any breaking of the symmetry due to the experimental geometry is neglected), the laterally and time integrating spin-ARPES experiment is not capable of distinguishing the orientation of macroscopic magnetization $M_i(t)$ of a fluctuating spin block $i$ at a time $t$, at and above $T_C$, i.e. one cannot tell if $M_i$ fluctuates only between $M_i$ and $-M_i$, or is randomly distributed along all three dimensional directions. However, if the ensemble originates from the crystalline solid, then the directional nature of SOC, in principle, allows to distinguish between the two cases. Therefore, further insight into the physics of the phase transition in EuO is obtained by considering the effects due to SOC which requires defining the crystal quantization axis (the direction of the magnetization $M$). SOC introduces a directional dependence of the FM band structure that leads to non-negligible corrections in momentum resolved experimental spectra [46] and related fundamental transport phenomena [47].

There are three types of directions in our work that should not be confused, and we define them as following, referring them to the EuO crystalline directions. First, our spin-APRES data is taken in normal emission geometry, therefore it probes states along the [001] Γ − X trajectory in the BZ. Secondly, the quantization axis of our Ferrum spin detector is along the [010] in-plane direction of the EuO film. Third, we assume that the easy axis of our sample is along the [111] direction, and at low temperatures the sample is magnetized with $M$ parallel or antiparallel to this direction. Our spin-resolved experiment probes the projection of $M$ onto the Ferrum spin quantization axis and all measurements are performed in remanence.

Let’s assume that $M_i(t)$ of a fluctuating spin block $i$ at $T \geq T_C$ is oriented along a generic random direction in the three-dimensional space. The result of the experiment is then the sum of photoelectrons from all the blocks $i$, each of them with the randomly oriented $M_i$. Such interpretation is justified e.g. by resemblance to Bloch domain walls below $T_C$.

Our spin-ARPES spectra were taken at normal emission, which for our geometry probes the [001] direction in the BZ due to the conservation of the parallel momentum. Therefore, in the following we focus exclusively on the band structure along this direction, with the result for two different magnetization directions presented in Fig. 2(a). The solid lines in Fig. 2(a) represent normal emission bands with the sample magnetized along [111], corresponding to our low-temperature spectra. With SOC included, changing the magnetization direction clearly renormalizes band dispersions, with lifted degeneracies and shifts of the order of 50-200 meV.

If spin blocks indeed exist above $T_C$, one can assume that above a certain temperature the $M_i$ in each spin-block $i$ is oriented randomly. Fig. 2(c-d) attempts to simulate the change of the spin integrated spectral shape due to SOC between 0 K and $T_C$. Plotted are partial densities of states (pDOS) projected on the [001] direction, i.e. the direction normal to the surface. We neglect phonon-related thermal broadening, although phonon effects might be significant near $T_C$ [48]. Figures 2(c)-(d) consider an easy axis along [111] at low temperatures, and an average over all possible $M$ directions at and above the $T_C$. We have approximated this average by taking only $M$ along high symmetry directions, in particular, we included 6 <001> directions (i.e. the [100], [010], [001], [100], [001], and [001]), 12 <110> and 8 <111> directions, making the pDOS along [001] depicted by magenta curves in Fig. 2(c)-(d). This demonstrates that the spectral renormalization due to influence of SOC within the fluctuating spin blocks regime is small but non-negligible. In particular, in the case of Eu 4f one can observe significant shifts of the spectral weight.

A more exact comparison should include the finite $k_z$ resolution of ARPES experiment [49], which will amplify the differences in Fig. 2(c)-(d). Also, one should take into
account all the different generic directions of $\mathbf{M}$, however, it is unlikely it introduces significant differences, and properly converging a calculation with $\mathbf{M}$ along a generic direction might be challenging due to dense k-point sampling requirement. Furthermore, we take each $\mathbf{M}$ with the same weight, while likely there exist an intermediate temperature range, where there is a preferential orientation of spin blocks, e.g. along various $<111>$ directions, however, we do not consider it here.

IV. EXPERIMENT

EuO thin films were grown in a molecular beam epitaxy (MBE) system with a residual gas pressure $p < 2 \times 10^{-10}$ mbar on a Cu(001) single crystal substrate. The films are epitaxial with EuO(001)||Cu(001), and $d = 25$ nm thickness resulting in a bulk-like electronic structure [50]. The LEED and RHEED patterns obtained on the EuO(001) films can be found in the Supplemental Material, Fig. S3. Stoichiometry of the EuO films was achieved by using the distillation method [51–54] and was confirmed by in-situ X-ray photoelectron spectroscopy (XPS) (Supplemental Material, Fig. S4). The spin polarization of the photoemitted electrons was reversing after the sample was remagnetized, confirming its ferromagnetic character (Supplemental Material, Fig. S5). The bulk-like Curie temperature of $T_C \approx 72$ K, was verified using vibrating sample magnetometry (VSM) measurements (Supplemental Material, Fig. S6).

High-resolution ARPES and spin-ARPES measurements were carried out in another ultra-high-vacuum (UHV) chamber ($p < 1 \times 10^{-10}$ mbar). Samples were transferred using a transportable UHV shuttle. After the vacuum transfer, the samples were annealed up to 300°C for 2 min to desorb surface contaminants. The spectra were taken using non-monochromatized He Iα resonance radiation with $h\nu = 21.22$ eV. The detailed discussion of the possible electronic transitions within the free electron final state model is presented in the Supplemental Information, Fig. S7. The energy resolution in the ARPES mode was 20 meV and in the spin-ARPES mode 50 meV. The parallel momentum resolution in spin-ARPES mode was $\leq 0.1\text{Å}^{-1}$. The spin-polarization of the photoelectrons was measured at normal emission with a FERRUM spin detector [55] that has a Sherman function $S = 0.29$. The FERRUM detector measures two spectra $I_+$ and $I_-$ for the target magnetized in opposite directions. Spin polarization is calculated as $P = (1/S)(I_+ - I_-)$, and in the conventional procedure the up/dn spectra are reconstructed as $I_{\uparrow(\downarrow)} = 0.5(I_+ + I_-)(1 \pm P)$.

We have measured two EuO films grown on different days, which we denote the "Sample A" and the "Sample B". Measuring EuO surfaces with laterally averaging technique such as ARPES is challenging due to the EuO surface reactivity and surface aging. Because of this, even though the ultra-high vacuum transfer between MBE and ARPES chambers has been made immediately

![Figure 3](image_url)

**FIG. 3.** (a): Set of temperature-dependent normal emission spectra normalized on the Eu 4f area from "Sample A". The legend shows chronological sequence of taking the spectra. (b): O 2p peak area from (a) with the same intensity scaling, but with binding energies aligned on the right (low binding energy) slope at half-maximum to allow comparison of the peaks widths and shapes. (c): the same as (b) but for Eu 4f. Temperature dependent positions of the Eu 4f and O 2p slopes are plotted in (d) and (e), while (f) shows their difference, the distance between the two peaks. (g): FWHM of the Eu 4f, taken on slightly smoothed spectra. Dashed lines in (d-g) show these values for 300K.
after the growth, flashing of the sample to 300°C before the measurement was needed to observe sharply dispersing bands.

**IV.1. Angle-resolved and spin-integrated photoemission**

Figure 3 shows the temperature-dependent set of normal emission spectra from "Sample A". Fig. 3(a) shows the temperature series of normal emission EDC spectra normalized on the Eu 4f peak. Note that in Fig. 1 we have used an initial energy scale, that is negative for occupied bands and referenced to the top of the 4f manifold, and in Fig. 3, as well as in subsequent Figs. 4, and 5 we use a binding energy scale referenced to the Fermi level measured on the metal plate in contact with the sample. The two scales are shifted with respect to each other, and similarly to Ref. [24] the theoretical energy separation between 4f and 2p manifold is too small, see also Fig. S7(a)-(b) of the Supplemental Material. This discrepancy does not influence our conclusions, and has been previously addressed by calculations with correlation effects included from first principles [33].

Chronologically, the 300K EDC was measured first, then the sample was cooled down to ∼ 30 K, and warmed up in steps as indicated in the legend of Fig. 3(a). The intensity ratio between O 2p and Eu 4f steadily grows over time, that is, depends mostly on the surface aging and not on the temperature. Therefore, the changes in the shape of the O 2p peak in Fig. 3(b) cannot be considered as being of a purely magnetic origin. Eu 4f spectra in Fig. 3(c) exhibit changes in the FWHM of the order of 70 meV, summarized in (g), with the general trend of the FWHM getting narrower at higher temperatures which suggests the bandwidth narrowing due to the Pauli localization with the increased magnetic disorder. This trend is in agreement with the theoretical calculations (Fig. 1), where the bandwidth is largest for the FM phase.

Although the separate sub-bands of the Eu 4f cannot be observed in our experiments, likely due to the correlation effects that tend to smear out spectral features (as shown in the context of ARPES e.g. for transition metals [56]), the effects of SOC summarized in Fig. 2(d) can at least partially account for minor changes observed in the experimental shape of the normal emission Eu 4f spectra in Fig. 3(c). In total DOS these changes would be negligible both for Eu 4f and O 2p since over the entire BZ the directional effects of SOC are averaged out, therefore, one can expect differences between momentum resolved (e.g. ARPES, spin-ARPES) and momentum integrating (e.g. optical spectroscopy, x-ray absorption) methods.

The reliable interpretation of O 2p FWHM is obscured by the strong spectral renormalization due to the aging. Nevertheless we provide the summary in Supplementary Fig. S10 (panel d8), which indicates the peak narrowing when warming up the sample, however, also a clear inconsistency with the RT width. Despite that, any narrowing in a series of spectra taken at incrementally increased temperatures might indicate a partial exchange splitting collapse in O 2p. Unfortunately, the aging issues prevent an unambiguous interpretation of these results.

Figs. 3(a) and (d-e) indicate binding energy shifts between the spectra, with the similar order of magnitude but different behavior as compared to the previous work [25]. The origin of this difference might be related either to the surface aging or to the band bending and the photovoltage between the Cu(001) and the EuO film. Such effect are difficult to quantify and they may influence the peak width analysis. However, the separation of the peaks in Fig. 3(f) is consistent with the Ref. [25], being larger for the FM phase.

Figure 4 compares RT and 40 K spin-integrated ARPES spectra from "Sample B" ("Sample A" shows nearly identical results). Chronologically, after the vacuum transfer and preparation by annealing, the RT spectrum in Fig. 4(a) was measured. Immediately after cooling to 40 K the spectrum in (c) was measured, and a further RT spectrum (e) was measured the next day after warming up the cryostat and another annealing cycle. Second derivative spectra indicate that near normal emission the O 2p exhibits two-peak structure at RT and four-peak structure at 40 K, in agreement with similar analysis used in the previous work [25]. The origin of these features can be found by inspecting the data from Fig. 3(a), where low temperature O 2p features exhibit shoulders on the high binding energy side. This behavior could be interpreted as a signature of the collapse of the exchange splitting, however, such an effect should lead to the constant area under the O 2p peak. Testing different ways of normalization of the spectra leads to the conclusion that the O 2p area does not remain constant, which supports the scenario of the surface contamination being responsible for the difference between RT and low temperature spectra.

**IV.2. Angle-resolved and spin-resolved photoemission**

To tackle the difficulties in interpreting standard spin-integrated ARPES, we turn to the spin-resolved version of this technique. Figure 5 shows a series of temperature-dependent spin-ARPES spectra from "Sample B". Figure 5(a) shows spin-resolved spectra for the sample in a ferromagnetic state at $T \simeq 40$ K, where the temperature-derived depolarization should be small. The O 2p manifold in Fig. 5(a) shows $2\Delta E_{ex} \simeq 0.3$ eV. Figures 5(a)-(d) reveal how the spin-resolved spectra evolve with the increasing temperature and (e)-(h) present the corresponding spin polarization distributions. It is evident that the spin-polarization in the photoemitted ensemble converges to 0 upon approaching $T_C \simeq 72$ K (Fig. 5(d) and (h)).

Due to the intra-atomic exchange splitting of the order of 10 eV the Eu 4f manifold is virtually 100 % spin polarized (see Supplementary Fig. S2), however, in Fig.
It is important to note, that a simple interpretation of the function of $S_{\text{eff}}$ is justified only assuming the fluctuating spin blocks. More general rules on spectral behavior when adjacent Eu 4f spins are not aligned would require theoretical modeling that takes into account the short range order \cite{3, 57}. Having that in mind, a fitting procedure was applied to the spectra shown in Figs. 5(i-k) (that were calculated using their respective different $S_{\text{eff}}$) to extract the $2\Delta E_{\text{ex}}$ of the O 2p bands assuming the scenario of spin blocks. We took advantage of the double peak shape of each spin channel, and used Voigt doublets for fitting. Figure 6 summarizes the temperature-dependent polarization in Eu 4f [Fig. 6(a)] and the results of the fitting procedure [Fig. 6(b)]. In Fig. 6(a) the Eu 4f spin polarization is presented as open squares, in comparison to the Brillouin function for the spin $7/2$, shown as a full line. Taking into account the experimental uncertainty, we can conclude that the experimental spin polarization vanishes at $T_C$, as predicted by the theoretical model.

Because of the fact that the ensemble spin polarization approaches zero when the sample temperature approaches $T_C$ [Fig. 6 (a)], the spin polarized spectra cannot be extracted at $T_C$. However, we are still able to fit the spin-integrated spectrum near $T_C$ (i.e. the sum of the two spin channels from Fig. 5(d)) using the doublet width and ratio parameters obtained from the spin-resolved spectrum of Fig. 5(k) measured below $T_C$, with the result shown in Fig. 5(l). As shown in Fig. 6(b) this indirect procedure indicates that the $2\Delta E_{\text{ex}}$ within the spin-block did not collapse at $T_C$. Details of the fitting procedure are described in the Supplemental Material (Section SVI), and Fig. S10 there shows similar analysis for the dataset from "Sample A" which further confirms our findings.

Figure 6(b) presents the values of the O 2p exchange splitting extracted from the fitted spectra. The crosses mark points obtained following the conventional analysis of the spin-resolved spectra, using the Sherman function of $S=0.29$. In this case, the exchange splitting approaches zero when the temperature reaches $T_C$. A significantly different picture emerges when one takes into account the existence of the spin blocks, and the analysis is performed using the effective Sherman function $S_{\text{eff}}$ (red dots). In this case, the exchange splitting remains nearly unchanged when the temperature approaches $T_C$, staying within the experimental uncertainty found from computing the 95% confidence intervals (gray areas). The results of the fitting of the spin-integrated spectra assuming $S_{\text{eff}}$ is shown for a comparison (black triangles). Similar results for the "Sample A" are presented in the Supplemental Information (Fig. S10). These results are the key finding of our work.
FIG. 5. Temperature-dependent spin-polarized photoemission of Eu 4f (located at \(\sim 2\) eV) and O 2p (at \(\sim 5\) eV) bands taken on "Sample B". (a-d): data from the spin-up (blue) and spin-down spectrum (red) of the spin-detector corrected by the Sherman function \(S = 0.29\). (i-k): respective calculated spin-polarizations. (l): Points show recalculated spectra (a-c) using the effective Sherman \(S_{\text{eff}}\) function adjusted to obtain the full spin polarization of Eu 4f. Cyan and magenta lines in (i-k) show Voigt doublet fits for the O 2p region. (l): Points show the sum of the up/dn spectra from (d), i.e. the spin integrated spectrum at 70K, together with the fit by Voigt doublets (see text for details). The gray area in (a) shows the binding energy region used to compute \(S_{\text{eff}}\), gray areas in (e-h) depict standard deviations for respective spin polarization curves, and gray line in (l) shows the fitted curve. Temperatures from left to right: 40K, 50K, 60K, 70K, temperature calibration accuracy 10 K.

V. DISCUSSION

Spin wave dispersions in EuO are known to be strongly renormalized when approaching \(T_C\), as shown in the early neutron scattering studies [58], which has led to the conjecture that above \(T_C\) magnons at all \(q\) are equally occupied, leading to a full magnetic disorder already at \(T_C\). On the other hand, further neutron scattering experiments near and above \(T_C\) indicate the existence of the short-range magnetic order [17]. It is also known that a non-vanishing susceptibility extends at least up to 1.4\(T_C\) [16]. Moreover, for itinerant magnets the existence of a short range order above \(T_C\) has been established [2, 5, 18, 19].

Two assumptions are important for our discussion. The first one is the adiabatic spin dynamics [5], i.e. the assumption that the response of the electronic system is much faster than the frequency of magnetic fluctuations. This the justified by noting that magnon energies, typically being less than 6 meV [58], are much smaller than the bandwidth of electronic dispersions. The second assumption is that the magnitude of the Eu 4f local moment is temperature independent. This is justified through the large intra-atomic exchange Eu 4f splitting of the order of 10 eV, small hybridization of the 4f with any other band visualized in Figs. 1 and S2), and small spectral weight renormalization depicted in Fig. 3(c)]. These properties of the Eu 4f levels make the EuO an ideal test system to verify the behavior of exchange splitting in the O 2p levels using spin-ARPES.

Clearly, our experimental spin-integrated ARPES data does not exhibit any spectacular renormalization near \(T_C\). The changes in the FWHM width of the O 2p and Eu 4f manifolds are of the order of 70 meV. We should point out that the data is likely affected by the surface aging effects since the EuO(001) surface does exhibit surface resonances in the O 2p so its properties might be affected by the contamination [the surface electronic structure of the relaxed EuO(001) surface is shown in the Supplemental Material of Ref. [59], while Refs. [60, 61] show the unoccupied part only]. Furthermore, a surprisingly small work function of EuO(001) [62, 63] suggests a high reactivity of this surface. From the comparison between FM and AFM band structures (Fig. 1), and from the arguments that in general the disorder localizes bands though the Pauli principle, the bandwidth of Eu 4f is expected to decrease with the magnetic disorder. Indeed, the width of Eu 4f is decreasing with higher temperatures, with the RT width being consistent with the 80K and 95K width, which suggests the magnetic origin. However, similar changes can be expected due to SOC when the spin blocks are assumed [Fig. 2(d)], as well as due to the surface aging.
tion accuracy of "Sample B" shown in Figs. 5(a-d). The temperature calibration accuracy of ±10 K is indicated with error bars.

(b) O 2p exchange-splitting as obtained by fitting the data of Fig. 5(a-d) (crosses) and of Fig. 5(i-k) (circles). Triangles show the result of the fitting of the exchange splitting in spin-integrated spectra. See text and Supplemental Material SV for details.

The spectral shape of O 2p is significantly affected by aging, with RT spectra (measured before cooling the sample) inconsistent with the 95K spectrum. The inconsistent spectral shape of the RT spectrum might be controlled by the temperature broadening. Predicted SOC effects appear too small to account for the observed experimental renormalization, especially in the higher binding energy portion which should be less affected by SOC. Spectra at lowest temperatures exhibit shoulders on high binding energy side, which leads to the 4-band structure. The routine procedure would mix the up and down exchange-split peaks, and, considering the broadening, lead to the conclusion of nearly vanishing exchange splitting that is in this case false.

The decoupling of the measured polarization and exchange splitting can be illustrated in the Eu 4f line that locally remains fully exchange-split up to RT and higher, while the standard analysis in Fig. 5(a)-(h) shows that its spin polarization vanishes at \( T_C \). Actually, exactly because of this, EuO spectra exhibit unique properties which allow testing the spin-block scenario using spin-ARPES up to the RT. Fully locally polarized Eu 4f can be used as a gauge for obtaining \( S_{eff} \) and recalculating the spin-ARPES spectra in an alternative way, Fig. 5(i)-(k) (and Supplemental S10). Surprisingly, according to such an analysis the spin splitting of the O 2p remains virtually constant for the temperatures up to 60 K, as shown for two different EuO films. The key finding is, that the change in the O 2p splitting established using \( S_{eff} \) [Fig. 6 (b)] does not follow the change in global magnetization that in EuO is characterized by the Brillouin function for spin 7/2 [Fig. 6 (a)]. These findings support the picture of the spin-block regime at least up to the \( T_C \).

A realistic description of the phase transition requires taking into account the nature of the magnetic short-range order [3, 57] and its impact on the space- and time-averaged spin-ARPES spectral function, an undertaking beyond the scope of this work. Since above \( T_C \) local properties are averaged in spin-ARPES, our analysis above \( T_C \) is based on the analysis of the peak widths and shapes. Theoretical work based on \( d - f \) model for the unoccupied Eu 5d manifold [32, 61] suggests that for many bands the collapse of the exchange splitting should be accompanied by a significant band broadening. This can also be intuitively understood from the fact that magnetic disorder breaks the crystal translation symmetry, and the momentum-resolved ARPES signal, which to the first approximation is the Fourier deconvolution of the supercell [64, 65], will be broadened by the disorder. Therefore, if the exchange splitting collapses, the expected peak width at higher temperatures should also include this broadening. However, there are multiple other possible sources of the line broadening in ARPES, such as the phonon
related effects, the final-state effects [66], and, for the dispersive bands, the wavevector broadening [67, 68].

Previous optical [26, 27], x-ray absorption (see Chap. 4 in [39]), and transport [29, 69] studies were interpreted as an evidence of the vanishing exchange splitting in the unoccupied bands near the CBM. The response of the conduction band is governed by the intra-atomic exchange between 4f and 5d electrons centered on the Eu sites within the d-f model [32]. It is important to note that even though the optical spectroscopy is sensitive to the band gap and does not directly probe the spin character of the bands. The temperature dependent changes in the optical spectra extend way beyond the $T_C$, suggesting the existence of the short range order. Strictly speaking, the fact that the absorption edge stabilizes above $\sim 100$ K [26] does not directly mean the exchange splitting in Eu 5d electrons has collapsed, but rather that the gap does not change anymore. Modern theoretical calculations predict $2\Delta E_{ex}$ in Eu 5d of the order of 0.8 eV (see [33] and Fig. 1), therefore the red shift of $\Delta E_{ex} = 0.4$ eV would indicate the full collapse, while the experiments indicate $\Delta E_{ex} = 0.25$ eV [26, 27]. This discrepancy has been previously discussed in [32], however, further progress in establishing the realistic exchange splitting at the bottom of the Eu 5d manifold and in taking into account the short range order seems necessary.

Furthermore, optical methods measure excitonic, and not electronic, gaps which might make them more sensitive to the macroscopic magnetization. Taking into account the relatively large EuO dielectric constant [70], excitons may have sizes much larger than the interatomic scale, while conduction band splittings are managed by the Eu 4f spin alignment on the scale of several interatomic distances.

The transport study [29] that has been performed using tunnel junctions with EuO thicknesses of 2-4 nm also indicates the existence of the short-range magnetic order above the $T_C$. Such thin films embedded into multilayers exhibit $T_C$ significantly lower than the bulk one [69] and the dimensionality is known to influence the character of magnetic order. Therefore, one can consider the results of Ref. [29] compatible with the spin-block conjecture for our bulk-like samples.

Previous work on a d-f model [32, 61] has addressed the temperature-dependent EuO exchange splitting in the Eu 5d part of the conduction band using the single-electron Green function formalism similar to the modern CPA schemes [15]. The model of Ref. [32] seems to decouple $2\Delta E_{ex}$ and band broadening, by definition leading to vanishing $2\Delta E_{ex}$ at $T_C$. Therefore, the picture of spin blocks and finite spin correlation length above $T_C$ is not included in this formalism. It is possible that the spectra at $T_C$ calculated in Ref. [32] are close to the case of the total magnetic disorder.

Simulating spin blocks (that is finite spin-correlation lengths) would require large unit cells, making ab-initio calculations prohibitive. Therefore, the picture that emerges from these considerations is that currently the complexity of the problem prevents realistic predictions on the temperature behavior of the exchange splitting in EuO above $T_C$. To circumvent this problem simulating larger unit cells, within e.g. tight binding models, may provide additional insight into the dependence of the exchange splitting on the spin correlation length.

**VI. SUMMARY AND OUTLOOK**

In summary, our experimental results of spin- and angle-resolved photoemission spectroscopy support the scenario of the short range magnetic order in EuO that exists across $T_C$ being accompanied by the non-vanishing exchange splitting of the O 2p states.

Our DFT calculations with a Hubbard $U$ term for FM, AFM-I, and AFM-II phases reveal a complex nature of the exchange splitting in the O 2p manifold, which depends on the arrangement of the 6 NN Eu 4f local moments. The bandwidths of the Eu 4f and O 2p manifolds decrease in AFM phases, indicating the expected Pauli localization. Calculations including SOC reveal renormalizations of the band dispersions which are particularly strong in Eu 4f bands and may account for their temperature-dependent changes.

Spin-integrated experimental spectra show dispersive features in the O 2p manifold indicating clean and well-ordered EuO(001) surfaces. Detailed temperature dependent analysis of the peaks widths reveals small narrowing of Eu 4f at higher temperatures. An unambiguous interpretation of this narrowing due to vanishing of the short range order is hindered by competing effects related to SOC within the fluctuating spin-blocks scenario. O 2p exhibit significant temperature-dependent renormalization, which, however, is also influenced by the surface aging effects, again preventing unambiguous interpretation. No dramatic temperature-dependent effects are observed between spectra above and below $T_C$, rendering the vanishing of the exchange splitting at $T_C$ unlikely.

Spin-ARPES spectra reveal the vanishing of the global magnetization at $T_C$, and in routine interpretation using constant Sherman function $S$ they indicate the vanishing of the exchange splitting in O 2p at $T_C$. Conversely, our alternative analysis assuming spin blocks and using the effective Sherman function $S_{eff}$ reveals virtually constant exchange splitting in O 2p up to 60 K. This result supports the scenario of fluctuating spin blocks being responsible for the phase transition.

Taking into account the above, it appears important to improve existing theoretical finite temperature models of EuO to account for the short range magnetic order above $T_C$ and relate it to the ARPES spectral function. From the experimental point of view it would be interesting to revisit the exchange splitting in the Eu 5d conduction band by methods that are able to probe unoccupied states directly, such as spin-resolved two-photon photoemission [19] or spin-resolved inverse-photoemission [71].
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