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Interference of spin states in resonant photoemission induced by circularly polarized light from magnetized Gd

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We have observed the spin-state interference by measuring the photoelectron spin polarization in the resonant preedge $4d \rightarrow 4f$ photoemission from magnetized Gd. The photoemission is induced by circularly polarized light which determines one preferential direction of electron spin orientation due to polarization transfer and spin-orbit interaction. Another direction perpendicular to the first one is determined by the target electron spin orientation connected with the target magnetization. We have measured the component of spin polarization perpendicular to those two directions which can only appear due to spin-state interference which implies coherence of the spin states produced by the two mechanisms of the photoelectron spin polarization.

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One of the basic principles of quantum mechanics is the principle of superposition. It asserts that if two states of a system are described by the wave functions Ψ_1 and Ψ_2, there exists an infinite number of other states described by the linear superposition $a\Psi_1 + b\Psi_2$ with a and b being some complex numbers.[1] Such a coherent superposition of quantum states often leads to interference effects observed in many experiments. Perhaps the most famous example is a two-slit experiment with electrons.[2] In this paper we deal with a coherent superposition of the two spin states of an electron which are produced by different mechanisms. Suppose that one mechanism produces an electron spin state oriented along z axis, represented by a spinor $\binom{1}{0}$, and another mechanism produces the spin state oriented along x axis described by a spinor $\binom{1}{1}$. If both states produced are added incoherently, the orientation of the spin polarization is inevitably in the xz plane. If, however, the states are added coherently, their linear superposition results in a new spin state $\binom{\alpha}{\beta}$ where α and β are complex numbers. It is easy to show that in this case the mean value $\langle \sigma_y \rangle$ of the y component of the spin is in general nonzero and equal to $2\,\mathrm{Im}(\alpha^* \beta)$. Thus as a result of the interference a spin polarization *perpendicular* to the original polarizations appears which could not appear in the incoherent case. This phenomenon is quite general and can appear in any branch of physics dealing with fermions. However, as far as we know, it was never observed experimentally. To investigate the spin-state interference, we study the spin polarization of photoelectrons in $4f$ photoemission (PE) spectra of metallic, ferromagnetic gadolinium in the vicinity of the $4d \rightarrow 4f$ giant resonance. The two preferential spin directions are defined by using the ferromagnetic order of the metal and the polarization transfer from circularly polarized radiation due to spin-orbit interaction.

Resonant PE is caused by a coherent superposition of the direct PE channel

$$\gamma + (4d^{10}4f^7)^8S_{7/2} \rightarrow (4d^{10}4f^6)^7F_J + \epsilon\ell \tag{1}$$

and the indirect excitation-autoionization channel

$$\gamma + (4d^{10}4f^7)^8S_{7/2} \rightarrow (4d^9 4f^8)X \rightarrow (4d^{10}4f^6)^7F_J + \epsilon\,\ell\ , \tag{2}$$

where $(4d^{10}4f^7)^8S_{7/2}$ is the ground state of the trivalent Gd and $\epsilon\ell$ represents the electron in the outgoing PE state. The intermediate resonant state X in our particular case is the $(4d^9 4f^8)^8D_{9/2}$ preedge resonance at the energy 138.8 eV. It decays by a super Coster-Kronig transition to the same final state as in the direct channel.

If the target is not polarized, then in the geometry of our experiment (see Fig. 1 and a description below) with the highly symmetric PE setup of normal incidence and normal emission, the spin polarization of photoelectrons can only have a component P_z parallel to the beam taken as z axis. Nonzero spin polarization of the direct photoelectrons can only appear due to the spin-orbit interaction if the fine structure components are resolved in the initial or/and the final state.[3] Since in our case the initial state is an S state and the final fine-structure components are not resolved, the direct process cannot produce any notable spin polarization for a nonmagnetized sample, which is confirmed by experiments (see below). In contrast, the resonant process which involves an intermediate state with a well-defined total angular momentum selectively enhances one of the fine-structure components. Therefore, a strong polarization of the resonant photoelectrons is expected which, for the present geometry, should be directed along the z axis. This polarization is determined by the mechanism of polarization transfer from the circularly polarized light to the photoelectron based on the spin-orbit interaction. In fact in our early experiment with paramagnetic Gd (Ref. 4) we have observed the polarization $P_z = 0.44 \pm 0.07$ at the considered resonance $E_\gamma \simeq 138.8$ eV

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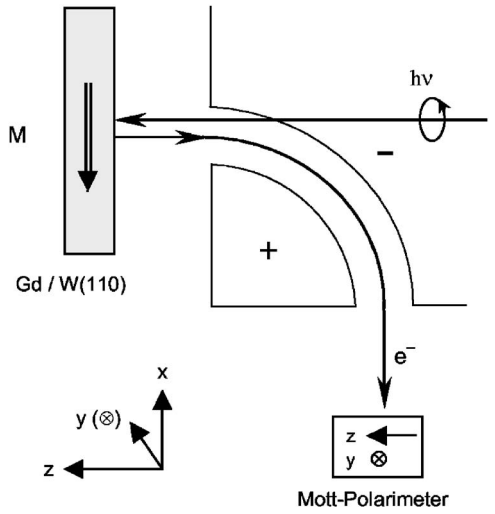

FIG. 1. Scheme of the experiment.

excited by circularly polarized radiation and $P_z \simeq 0.04$ for off-resonance emission at photon energy $E_\gamma = 130$ eV.

If the spins of the $4f$ electrons are oriented in a magnetized sample due to exchange interaction, this initial orientation is conserved even in the direct photoemission by unpolarized or linearly polarized light.[5] In our geometry (see Fig. 1) the magnetization is perpendicular to the photon beam and the electron spin polarization is oriented along the x axis. Thus separately the two mechanisms of spin orientation, polarization transfer in the resonant excitation and polarization of the target due to magnetization, produce spin polarized photoelectrons with the polarization vector in the z and in the x direction, respectively. Being coherently superposed these spin states give rise to the P_y component perpendicular to the xz plane detected in our experiment.

The experiment was carried out at the planar helical undulator beamline UE 56/2-PGM1 at BESSY (Berlin) using the apparatus utilized in the preceding experiment on paramagnetic Gd.[4] Figure 1 shows a scheme of the setup. The (circularly) polarized radiation from the beamline hits the target in normal incidence. Electrons emitted normal to the target surface pass a 90° spherical-field electron spectrometer (energy resolution <0.7 eV). The spectrometer is followed by a spherical-field Mott polarimeter with four counters allowing simultaneous determination of two polarization vector components, P_z along the incident beam and P_y perpendicular to both the beam and the magnetization vector $\vec{M}$ (see Fig. 1). Apparatus related asymmetries are eliminated by combining cycles with reversed helicity of the radiation.

The crystalline Gd films were grown onto a W(110) crystal surface. The W crystal was mounted on a manipulator allowing an azimuthal rotation of about 100° in the surface plane and cooling by liquid He. For temperature monitoring, a W-5%Re/W-26%Re thermocouple[6] was fixed to the crystal. For cleaning, the W crystal was heated in front of a O_2 doser up to 1400 K followed by repeated flashing to ~2200 K.

Well degassed Gd (99.99%) (Ref. 7) was evaporated from a Mo crucible heated by electron bombardment. During evaporation the W crystal was kept at 30–50 °C. The evaporation rate and time were chosen to get an approximately 10 nm thick Gd film in about 1.5 min, monitored by a quartz microbalance. During evaporation the pressure rose to $(1–2)\times10^{-10}$ mbar and fell down to the base pressure $<5\times10^{-11}$ mbar shortly after stopping evaporation. After evaporation, following the recipe given in Ref. 8, the film was tempered for 2.5 min at 700 K. Well-annealed films show the hexagonal low energy electron diffraction (LEED) pattern of Gd(0001). The Gd films cooled to about 80 K were magnetized along their easy direction of magnetization $[11\bar{2}0]$, Ref. 9, by applying a pulsed magnetic field of a solenoid. The magnetization was proved by rotating the crystal azimuthally by 90° to get the easy direction along the y direction in Fig. 1 and measuring the spin polarization of the $4f$ electrons excited by linearly polarized radiation of 148.5 eV (position of the main resonance; see Fig. 2 upper panel) in the Mott polarimeter. The resulting spin polarization was $(52\pm8)\%$ fitting well to magnetic circular dichroism results[10,11] and the spin polarization given for a Gd monolayer on Fe in Ref. 5.

Our experimental results are shown in Fig. 2. The measurements show a large P_z component (up to 44%) in agreement with the earlier experiment[4] and reveal a significant coherence effect with a spin polarization P_y varying from $P_y \sim -13\%$ to $P_y \sim 5\%$ within a photon energy interval of about 0.4 eV on the low energy side of the resonance. Both components strongly vary across the resonance revealing a typical resonant behavior in the considered energy region. It may be parametrized using a Fano-type approach to the vector correlation parameters in resonant photoionization.[12,13] In Fig. 2 we show a fit of the polarization data by the parametric expression suggested in Ref. 13. We note that the parameters which determine the position and the width of the resonant curves for polarization must be the same for P_y and P_z components.

In order to understand the different behavior of the P_y and P_z components of spin polarization quantitatively we have made *ab initio* calculations based on the multiconfigurational Dirac-Fock (MCDF) approximation using the density matrix and statistical tensor formalism.[14] The photoionization amplitudes for the direct channel as well as the photoexcitation and autoionization amplitudes for the resonant channel have been calculated using the program package RATIP.[15] The wave functions for the initial, intermediate, and final states of Gd are calculated within the MCDF approach using the GRASP92 program.[16] Both initial and final state configuration interactions are taken into account within the configuration space chosen according to the nominal nonrelativistic configurations describing the states. In such approximation the calculated spectrum of resonances agrees rather well with the experiment, but it needs to be shifted by 1.5 eV in order to reproduce the experimentally observed resonance energies. The calculated intensities of the polarized electrons for the fine-structure components of the final state have been summed up to the total intensity ignoring the fine-structure splitting.

The results of our calculations are shown in Fig. 3. In general, good agreement is found between the calculated P_z and P_y components across the resonance and the measured

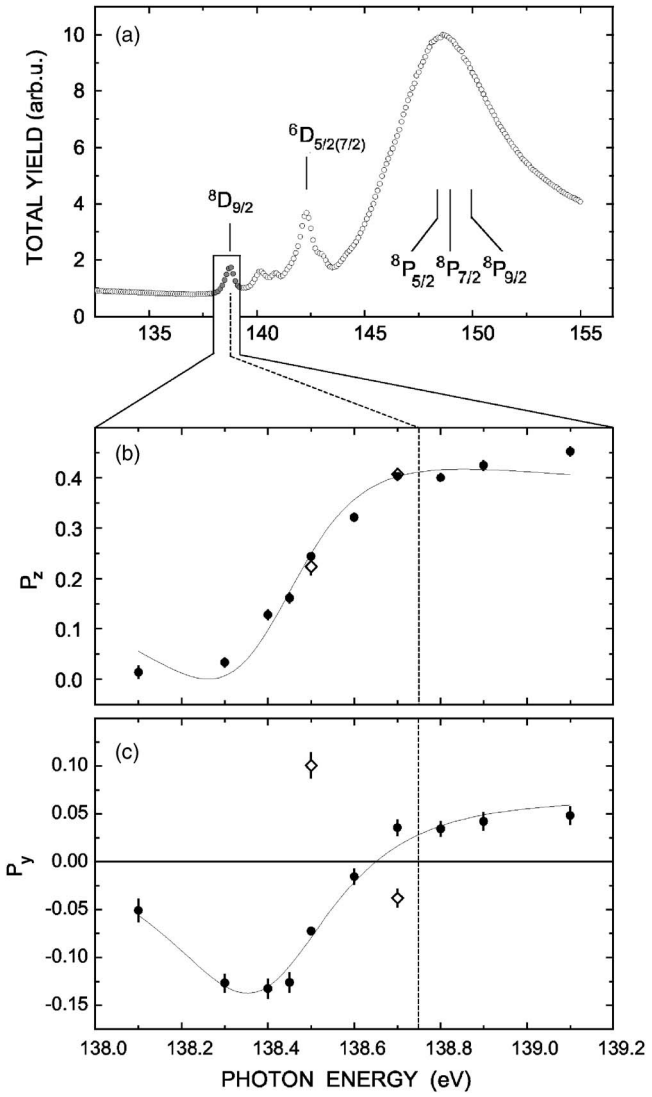

FIG. 2. (a) Total-yield spectrum of photoelectrons measured in the vicinity of the Gd $4d \rightarrow 4f$ resonance. Spin polarization components (b) P_z and (c) P_y measured across the first preedge peak at $E_\gamma = 138.75 \pm 0.1$ eV. The curves show the best fit for the parametrization suggested in Ref. 13. The vertical dashed lines show the position of the peak maximum. Black points are the results obtained with the magnetization directed as indicated in Fig. 1. Open diamonds show the results obtained with the reversed magnetization. The error bars give the single statistical errors connected with counting. The polarization scale contains a relative uncertainty of ±15% due to the uncertainty of the Mott-polarimeter calibration.

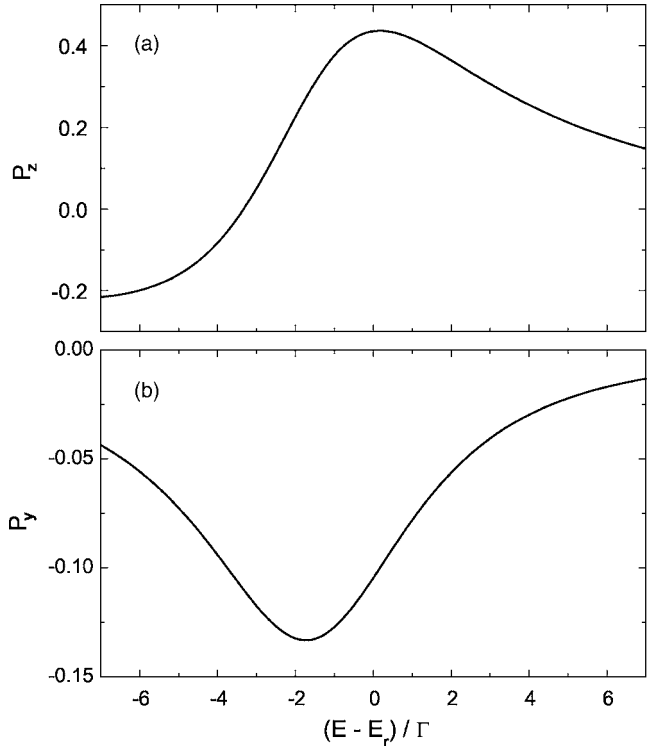

FIG. 3. Calculated photoelectron spin polarization components (a) P_z and (b) P_y in the vicinity of the first preedge resonance as a function of the photon energy. The energy is counted from the position of the resonance (E_r) in units of the resonance width (Γ).

data, although some of the features, like zero crossing in the P_y dependence, are not reproduced by the calculations. Overall, however, this is not surprising by taking into account the complexity of the problem owing to the open f-shell configuration of the initial, intermediate, and final states. We note that in the calculations there are two parameters: the degree of the spin orientation in the initial state and the width of the resonance which should be somehow estimated. To avoid the latter problem we present the calculated polarization (see Fig. 3) as a function of the reduced energy in units of the resonance width. The degree of orientation enters as a factor in the final expression for P_y; therefore, we simply normalized our curve to the maximal (modulus) experimental value of the P_y component. Practically the same result can be obtained if the degree of orientation is estimated from the experiment with the linearly polarized light mentioned above. Note that this parameter does not influence the P_z component; therefore, a good agreement between the calculation and experiment is achieved here without any fitting parameters. According to our analysis the change of sign of magnetization (P_x) changes the sign also in the P_y component, but does not change the sign of the P_z one. This is confirmed in the experiment (see Fig. 2 where the results for the reversed magnetization are shown by open diamonds).

In conclusion, we have measured the component of the photoelectron spin polarization perpendicular to the plane defined by the target magnetization and the helicity of the radiation. In this way we have demonstrated that the spin states produced by two different mechanisms—photoemission from a magnetized target and polarization transfer due to spin-orbit interaction—are coherent and therefore interfere with each other in going across the resonance. Our *ab initio* calculations qualitatively agree with the experimental data and clearly confirm this interpretation. The phenomenon of spin-state interference revealed by our experiment and thus of the rotation of the spin polarization vector out of the plane determined by the two original spin directions is of general importance for any resonant photoemission of ferromagnetic solids by use of circularly polarized light, since spin-orbit interaction is always present and

needed to couple the magnetic spin orientation to the lattice. Due to the interference nature of the spin rotation this phenomenon is very sensitive to the details of the resonant photoemission and may be useful in its study.

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