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Spin-polarized positron annihilation spectroscopy for spintronics applications

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Abstract. Spin-polarized positron annihilation spectroscopy will be useful in studying spintronics materials. Here, we summarize some fundamental aspects of this method for the future spintronics studies.

1. Introduction

Spin-polarized positron annihilation spectroscopy (SP-PAS) has so far been used for studying the polarized band structures of bulk ferromagnets [1–16]. Considering the facts that important spin-related phenomena such as magnetoresistance, spin current, spin accumulation and so on occur near surfaces/interfaces and spin device metarials such as spin injection electrodes are utilized in thin fims, by developing low-energy spin-polarized positron beam, SP-PAS will be further applied to the spintronics field. As for the sensitivity of positrons to vacancy defects, SP-PAS will also be an important tool to study vacancy-induced magnetism.

From the above viewpoints, we have been developing spin-polarized positron beams. We performed SP-PAS experiments with the Doppler broadening of annihilation radiation (DBAR) measurements on some ferromagnets. For studying surface spin phenomena, we revive the spin-polarized positronium (Ps) annihilation method based on the DBAR measurements, of which the original principle was established by the Michigan group using annihilation lifetime measurement [17]. In this paper, we briefly report the above two experiments.

2. SP-PAS for ferromagnetic systems

The positron-electron momentum densities for the *ith* majority (spin-down (↓)) and minority (spin-up (↑)) bands, $\rho_i^{\downarrow(\uparrow)}(\boldsymbol{p})$, of a ferromagnet are given by

$$\rho_i^{\downarrow(\uparrow)}(\boldsymbol{p}) = \left| \int e^{-i\boldsymbol{pr}} \Psi_+(\boldsymbol{r}) \Psi_i^{\downarrow(\uparrow)}(\boldsymbol{r}) \sqrt{\gamma[n_-(\boldsymbol{r})]} d\boldsymbol{r} \right|^2, \tag{1}$$

where $\Psi_+(\boldsymbol{r})$ is the positron wave function, $\Psi_i^{\downarrow(\uparrow)}(\boldsymbol{r})$ is the electron wave function and $\gamma[n_-(\boldsymbol{r})]$ is the enhancement factor. According to Berko [5,6], DBAR spectra (convolutions of apparatus resolution function and 1D-ACAR spectra) under the fields parallel and anti-parallel to the direction of motion of the positrons are given by

$$N_\pm(p_z) = \frac{\lambda_S}{4} \sum_{i=1}^{occ} \left[\frac{(1\pm P) N_i^{\downarrow}(p_z)}{\lambda^{\Uparrow}} + \frac{(1\mp P) N_i^{\uparrow}(p_z)}{\lambda^{\Downarrow}} \right], \tag{2}$$

where summation is over all occupied states, $\lambda_S = 4\pi r_e^2 c$ (r_e: the classical electron radius), P is the longitudinal polarization of positrons, $N_i^{\downarrow(\uparrow)}(p_z)$ is the double integral of $\rho_i^{\downarrow(\uparrow)}(\boldsymbol{p})$ and $\lambda^{\Uparrow(\Downarrow)}$ is the total annihilation rate of spin-up (spin-down) positrons:

$$\lambda^{\Uparrow(\Downarrow)} = \frac{1}{2}\sum_{i=1}^{occ}[\lambda_S w_i^{\downarrow(\uparrow)} + \lambda_T(w_i^{\downarrow(\uparrow)} + 2w_i^{\uparrow(\downarrow)})]. \tag{3}$$

Here, $\lambda_T = \lambda_S/1115$ and $w_i^{\downarrow(\uparrow)}$ is the overlap integral between the positron and ith majority (minority) spin band wave functions ($= \int\int\int \rho_i^{\downarrow(\uparrow)}(\boldsymbol{p})d\boldsymbol{p}$). Equation 2 suggests that $N_+(p_z)$ and $N_-(p_z)$ exhibit the characters of more majority and more minority spin bands, respectively. Their difference is given by

$$N_+(p_z) - N_-(p_z) = \frac{\lambda_S P}{2}\sum_{i=1}^{occ}\left[\frac{N_i^{\downarrow}(p_z)}{\lambda^{\Uparrow}} - \frac{N_i^{\uparrow}(p_z)}{\lambda^{\Downarrow}}\right]. \tag{4}$$

The summation of partial differential spectra $(N_i^{\downarrow}(p_z) - N_i^{\uparrow}(p_z))$ is given by

$$\begin{aligned}\sum_{i=1}^{occ}[N_i^{\downarrow}(p_z) - N_i^{\uparrow}(p_z)] &\propto \Delta N(p_z) + P\frac{\lambda^{\Uparrow} - \lambda^{\Downarrow}}{\lambda^{\Uparrow} + \lambda^{\Downarrow}}\Sigma N(p_z) \\ &\approx \Delta N(p_z) + P^{3\gamma}\Sigma N(p_z),\end{aligned} \tag{5}$$

where $\Delta N(p_z) = N_+(p_z) - N_-(p_z)$, $\Sigma N(p_z) = N_+(p_z) + N_-(p_z)$ and $P^{3\gamma} = (N_+^{3\gamma} - N_-^{3\gamma})/(N_+^{3\gamma} + N_-^{3\gamma})$, where $N_\pm^{3\gamma}$ denotes the three-photon annihilation intensities in positive and negative fields. Therefore, either through eq.(4) or eq.(5), one can extract the components related to magnetic electrons.

From eq. (3), the positron annihilation lifetime spectrum of a ferromagnet is composed of two different lifetime components even if P=0. The lifetime spectra in positive and negative field are given by

$$L_\pm(t) = \frac{\lambda_S}{4}\sum_{i=1}^{occ} w_i^{\downarrow}(1\pm P)exp(-\lambda^{\Uparrow}t) + \frac{\lambda_S}{4}\sum_{i=1}^{occ} w_i^{\uparrow}(1\mp P)exp(-\lambda^{\Downarrow}t). \tag{6}$$

Figure 1(a) shows the experimental set-up of DBAR measurement in magnetic field. Figure 1(b) shows the differential DBAR spectra obtained for polycrystalline Fe, Co and Ni samples under magnetic field using a ^{68}Ge-^{68}Ga source [18]. These spectra exhibit that $3d$ electrons having broader momentum distribution are responsible for polarization effects. The magnitude of field-reversal asymmetry reflects the difference of magnetization of these ferromagnets.

3. SP-PAS for surface spin systems

A fraction of positrons implanted near a metal surface are emitted as Ps by picking up surface electrons near the Fermi level. Ps annihiltion characteristics depends on the relative direction of positron and electron spin polarizations. The energy spectrum and annihilation lifetime of three-photon annihilation of spin-triplet Ps are separated from those of two-photon annihilation due to spin-singlet Ps and free positron annihilation. To detect spin-polarized electrons through Ps annihilation, it is convenient to observe the three-photon annihilation event. The Michigan group first demonstrated this experiments based on lifetime measurement [17]. We show below that the similar experiment is possible with DBAR measurement.

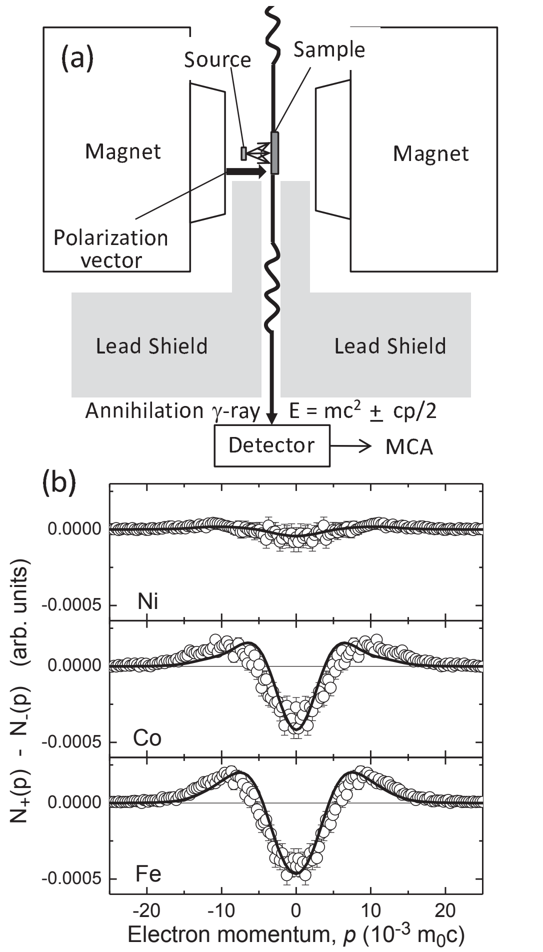

Figure 1. (a)Experimental set-up of DBAR measuerments under magnetic field. (b)Differential DBAR spectra obtained for polycrystalline Fe, Co and Ni samples under magnetic fields of ± 1 T.

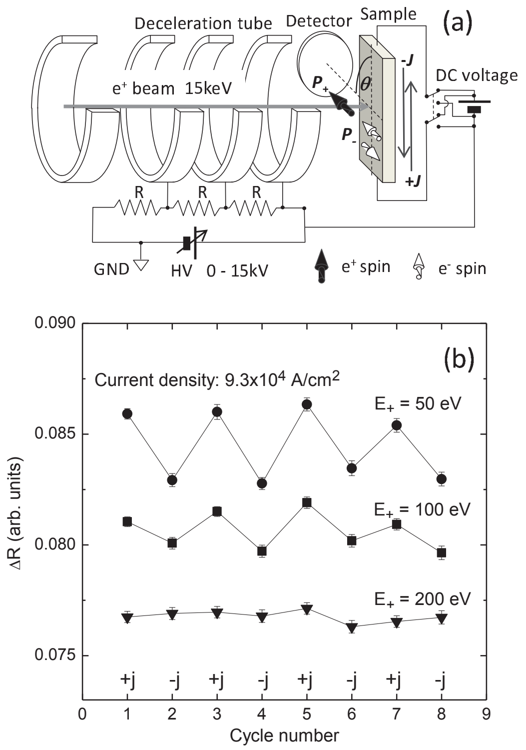

Figure 2. (a)Experimental set-up for detecting surface spin polarization under direct current. (b)Current reversal (+j ↔ -j) oscillation of $\triangle R$ due to spin-triplet positronium obtained for a Pt thin film surfaces at different incident positron energies (E_+).

Fractions of each spin state of Ps are given by

$$F_{|00\rangle} = (1 - P_+P_-cos\varphi)/4, \tag{7}$$

$$F_{|10\rangle} = (1 - P_+P_-cos\varphi)/4, \tag{8}$$

$$F_{|11\rangle} = (1 + P_+ + P_-cos\varphi + P_+P_-cos\varphi)/4, \tag{9}$$

$$F_{|1-1\rangle} = (1 - P_+ - P_-cos\varphi + P_+P_-cos\varphi)/4, \tag{10}$$

where the subscript $\mid Sm\rangle$ of F denote total spin S and magnetic quntum number m, $P_\pm$ is the positron and electron polarizations, respectively, and φ is the angle between the positron and electron polarization directions. The total three-photon annihilation fraction is given by

$$F^{3\gamma}_{Ps} = \epsilon(1)(F_{|11\rangle} + F_{|1-1\rangle}) + \epsilon(0)F_{|10\rangle}, \tag{11}$$

where $\epsilon(1)$ and $\epsilon(0)$ are the detection efficiencies of annihilation photons from m=±1 and m=0 states, respectively [19]. When the positron and electron polarization directions are the same, the $|11\rangle$ state is preferentially formed and the fractions of the other states are relatively lower, resulting in the increase of $F^{3\gamma}_{Ps}$. Conversely, when the positron and electron polarization directions are opposite, the fractions of the $|11\rangle$ and $|1-1\rangle$ states are relatively lower as compared with the $|10\rangle$ and $|00\rangle$ states. Hence, $F^{3\gamma}_{Ps}$ decreases. Ps three-photon annihilation is traditionally characterized by a quantity R [20],

$$\begin{aligned} R &= (T-U)/U \\ &= \frac{(1-F^{3\gamma}_{Ps})R_0 + F^{3\gamma}_{Ps}R_1U_1/U_0}{1-F^{3\gamma}_{Ps} + F^{3\gamma}_{Ps}U_1/U_0}, \end{aligned} \tag{12}$$

where T and U denote the area under the entire intensity curve and under the two-photon annihilation spectrum and the subscripts 0 and 1 denote the cases of 0% and 100% positronium emission, respectively. Although R is a complicated function of $F^{3\gamma}_{Ps}$, for a small $F^{3\gamma}_{Ps}$, $\triangle R$=R-R_0 is approximately proportional to $F^{3\gamma}_{Ps}$. Thus, by using Eqs.(8) to (12), the asymmetry of $\triangle R$ upon spin flip ($+P_- \leftrightarrow -P_-$) is

$$A = \frac{\triangle R(+P_-) - \triangle R(-P_-)}{\triangle R(+P_-) + \triangle R(-P_-)} = \frac{2\epsilon(1)-\epsilon(0)}{2\epsilon(1)+\epsilon(0)}P_+P_-cos\varphi. \tag{13}$$

Therefore, from the observation of $\triangle R$ by changing the relative direction between positron and electron spin polarizations, polarized electrons are detected.

Figure 2(a) shows the experimental set-up for the current-induced spin polarization on metal surfaces. Figure 2(b) shows the $\triangle R$ obtained for a Pt thin film (30 nm thick) under direct current as a function of successive current reversal at different incident positron energies. From this result, excess electron spins are accumulated on the Pt surface under direct current.

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