# Quadrupole interaction of positronium in $\alpha$-quartz

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Effective positronium $[\mathrm{Ps}=(e^+e^-)]$ quadrupole interaction in $\alpha$-quartz has been observed using the Zeeman effect on Ps. In order to reduce sample dependence and to achieve a good resolution, the two-dimensional angular correlation of annihilation radiation technique has been used. The quadrupole coupling constant $d$ in $\alpha$-quartz has been determined to be $(3.0\pm0.9)\times10^{-5}$ eV.

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It is well known that all hydrogen-like atoms consisting of electrically positive and negative particles with different masses possess a quadrupole moment due to the hyperfine interaction. This is predicted by Baryshevsky and Kuten[1] and confirmed by the discovery of the anisotropy of the hyperfine splitting of muonium(Mu) in $\alpha$-quartz.[2]

Positronium (Ps), which is the lightest H-like atom of an electron and its antiparticle, a positron, does not, in principle, have any multipole moments in vacuum, since the masses of the particle and antiparticle are the same. As a consequence, "vacuum" Ps is a totally CPT-invariant quantum system with identically zero multipole moments. However, Baryshevskii[3] predicted that Ps in material might possibly have an *effective* quadrupole moment since the effective masses of the electron and the positron are different. Bondarev and Kuten[4] developed a theoretical model for effective quadrupole moment $Q_{\mathrm{Ps}}$ and showed that it takes the form

$$Q_{\mathrm{Ps}}=\frac{m_e-m_p}{m_e+m_p}\langle 3z^2-r^2\rangle, \tag{1}$$

where $m_e$ and $m_p$ are the effective masses of the electron and the positron, respectively, and $\langle\cdots\rangle$ denotes the quantum mechanical averaging over the exact triplet ground state wave function taking the electron-positron hyperfine interaction into account. They also suggested that when Ps is in an oriented crystal, $Q_{\mathrm{Ps}}$ can be detected, in principle, through the measurement of the anisotropy of the fraction of Ps atoms which self-annihilate in a magnetic field. In the case where the oriented crystal is $\alpha$-quartz, the anisotropy was estimated theoretically to be $\sim0.1\%$ (Ref. 4) or $\sim1\%$,[5] depending on initial assumptions.

Gessmann *et al.*[6] measured the anisotropy of magnetic quenching of Ps in $\alpha$-quartz by using the positron spin relaxation technique and reported an anomalously large anisotropy of the hyperfine tensor of Ps ($A_{zz}/A_{xx}\approx1.93$). However, ambiguity remains since they used the Doppler broadening method whose resolution is relatively poor. In the present study we investigate the Ps quadrupole interaction by using the two dimensional angular correlation of annihilation radiation (2D-ACAR) method.

Ps has two ground states: *para*-positronium ($p$-Ps) and *ortho*-positronium ($o$-Ps). In vacuum and without the presence of a magnetic field, $p$-Ps and $o$-Ps annihilate into two and three $\gamma$-quanta with mean lifetimes of 125 ps and 142 ns, respectively. In condensed matter, however, most of the o-Ps atoms undergo pickoff annihilation, i.e., $2\gamma$ annihilation of the positron bound in Ps with a foreign electron from its environment.

If Ps has a quadrupole moment, the Ps spin Hamiltonian $H$ in a static magnetic field $\mathbf{B}$, is written in atomic units as[4]

$$H=\omega\mathbf{S_e}\cdot\mathbf{S_p}-2\mu_B(\mathbf{S_e}-\mathbf{S_p})\cdot\mathbf{B}+\frac{Q_{\mathrm{Ps}}}{4}Q_{ik}V_{ik}, \tag{2}$$

where $\mu_B$ is the magnetic moment of an electron, $Q_{ik}=F_iF_k+F_kF_i-(2/3)F(F+1)\delta_{ik}$ is the symmetric Cartesian tensor, $\mathbf{F}=\mathbf{S_e}+\mathbf{S_p}$ is the total angular momentum operator, $V_{ik}$ is the electric field gradient (EFG) tensor at the center-of-mass of Ps ($V_{ik}=V_{ki}$, $\Sigma_iV_{ii}=0$), and $\omega=\kappa\omega_0$ is the hyperfine splitting frequency of the Ps ground state in the crystal ($\kappa$ is the relative contact density of Ps and, in vacuo, $\omega=\omega_0=8.41\times10^{-4}$ eV). Considering the quadrupole term as a perturbation, four eigenstates $|n\rangle$ ($n=0,1,2,3$) of Ps with the Hamiltonian in Eq. (2) are given by

$$|n\rangle=\sum_{F,M}c_n^{(FM)}|FM\rangle, \tag{3}$$

where

$$c_{0,1}^{(00)}=\pm C_{0,1},\quad c_{0,1}^{(10)}=\cos\vartheta C_{1,0},$$

$$c_{0,1}^{(11)}=-\frac{\sin\vartheta}{\sqrt{2}}e^{-i\varphi}C_{1,0},\quad c_{0,1}^{(1-1)}=\frac{\sin\vartheta}{\sqrt{2}}e^{i\varphi}C_{1,0},$$

$$c_{2,3}^{(00)}=0,\quad c_{2,3}^{(10)}=\frac{\sin\vartheta}{\sqrt{2}}e^{i\varphi},\quad c_{2,3}^{(11)}=\pm\frac{1\pm\cos\vartheta}{2},$$

$$c_{2,3}^{(1-1)}=\pm\frac{1\mp\cos\vartheta}{2}e^{i2\varphi}, \tag{4}$$

 

$$C_0=\frac{1}{\sqrt{1+y_q^2}},\quad C_1=\frac{y_q}{\sqrt{1+y_q^2}},$$

$$y_q=y\sqrt{1+\frac{d}{\omega}(3\cos^2\theta-1+\eta\sin^2\theta\cos2\varphi)/2\sqrt{1+x^2}},$$

$$y=\frac{\sqrt{1+x^2}-1}{x},\quad x=\frac{4\mu_B B}{\omega}. \tag{5}$$

$|FM\rangle$ stands for the Ps eigenstate with total momentum $F$ and its $z$-projection $M$ in the absence of a magnetic field, $\theta$ and $\varphi$ are the polar and the lateral angles characterizing the tilt of **B** in the system of the EFG tensor principal axes, $d=Q_{Ps}V_{zz}$ is the quadrupole coupling constant of Ps and $\eta=|(V_{xx}-V_{yy})/V_{zz}|$ is the asymmetry parameter of the EFG tensor. Equations (3) and (4) with (5) show that the mixing of $|1M\rangle$ and $|00\rangle$ in the presence of a magnetic field does not depend only on the magnetic flux density, but also on the relative orientation of the magnetic field and the principal axes of the crystal lattice.

The mixing of the states enhances the intensity of the self-annihilating Ps. The fractions of $|1\rangle$ (*ortho*-like Ps) and $|0\rangle$ (*para*-like Ps) created from the spin polarized positrons in the magnetic field are[7]

$$F_1=\frac{F_{\mathrm{Ps}}}{8(1+y_q^2)}[(1+y_q)^2(1-P)+(1-y_q)^2(1+P)], \tag{6}$$

$$F_0=\frac{F_{\mathrm{Ps}}}{8(1+y_q^2)}[(1-y_q)^2(1-P)+(1+y_q)^2(1+P)], \tag{7}$$

where $F_{\mathrm{Ps}}$ is the Ps formation probability and $P$ is the projection of the incident positron polarization vector on the magnetic field direction at the instant of Ps formation. Thus, the fraction of the Ps atoms which self-annihilate into $2\gamma$ in the material is given by

$$I_{\mathrm{Ps}}(B)=\frac{\kappa\gamma_s y_q^2}{\Gamma_1(B)(1+y_q^2)}F_1+\frac{\kappa\gamma_s}{\Gamma_0(B)(1+y_q^2)}F_0, \tag{8}$$

where $\Gamma_1(B)$ and $\Gamma_0(B)$ are the total annihilation rates of the *ortho*-like and *para*-like Ps atoms, respectively, in the material:

$$\Gamma_1(B)=\kappa\frac{\gamma_t+y_q^2\gamma_s}{1+y_q^2}+\gamma_{po}, \tag{9}$$

$$\Gamma_0(B)=\kappa\frac{y_q^2\gamma_t+\gamma_s}{1+y_q^2}+\gamma_{po}. \tag{10}$$

Here $\gamma_t$ and $\gamma_s$ are the self-annihilation rates of *o*-Ps and *p*-Ps in vacuum, respectively, and $\gamma_{po}$ is the Ps pickoff annihilation rate in the material. Under the condition of $y_q^2\ll1$, $I_{Ps}(B)$ is given by[5]

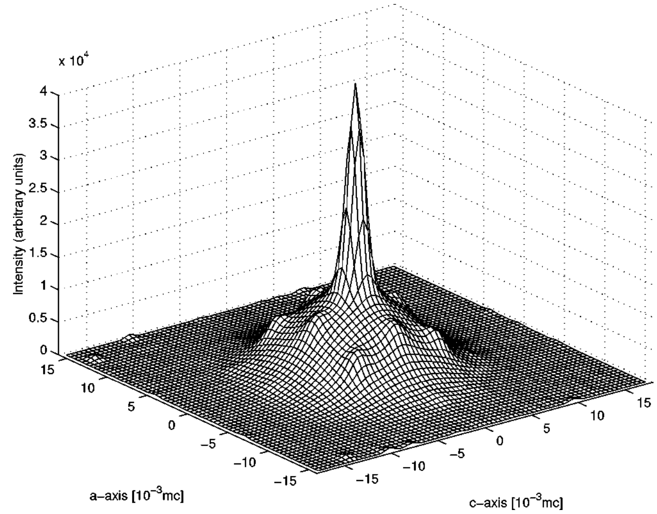


FIG. 1. 2D-ACAR spectrum for $\alpha$ quartz.

$$I_{\mathrm{Ps}}(B,\theta,\varphi)\approx\Xi(B,\theta,\varphi)\cdot I_{\mathrm{Ps}}(0), \tag{11}$$

$$\Xi(B,\theta,\varphi)=\frac{1+2\frac{\Gamma_0(0)}{\Gamma_1(0)}y_q^2+2Py_q}{1+\frac{\Gamma_0(0)}{\Gamma_1(0)}y_q^2}. \tag{12}$$

The anisotropy of $I_{\mathrm{Ps}}(B,\theta,\varphi)$ thus depends on the possible Ps quadrupole interaction.

$I_{\mathrm{Ps}}(B,\theta,\varphi)$ was measured using the 2D-ACAR method with the same sample to avoid possible sample dependence. Instead of changing the direction of the magnetic field, the sample was simply rotated. The movement of the sample around the axis parallel to the sample-detector line yields the rotated but identical Ps momentum projections with the changed magnetic field direction. It is to be noted that if we use the 1D-ACAR method, the rotation of the sample leads to a change of direction of the projection of the Ps momentum distribution. Therefore the use of the 2D-ACAR method resolves this problem.

The details of the 2D-ACAR system are described elsewhere.[8] The sample was cut to a size of $\sim3$ mm $\times3$ mm$\times10$ mm. The long edge was parallel to the $\hat{a}$ axis, and one of the short edges was parallel to the $\hat{c}$ axis, making the other short edge parallel to the $\hat{a}'$ axis of the reciprocal lattice. It was placed on the sample holder such that the long edge of the sample was along the direction of the detected $\gamma$ rays. The orientation of the crystal lattice was determined to be within 1° using x-ray diffraction before setting the sample. The magnetic field was applied perpendicular to the direction of the detected $\gamma$ rays. The $\hat{c}$ axis was set parallel and perpendicular to the magnetic field by rotating the sample.

Using a source whose intensity was $\sim15$ mCi, two spectra were measured in each scan in magnetic fields of 1.25 and $-0.25$ T. The sample was rotated after every four scans.

Figure 1 shows the 2D-ACAR spectrum for $\alpha$ quartz. One sharp peak is seen at the center of the spectrum representing the self-annihilation of Ps. There are also small satellite

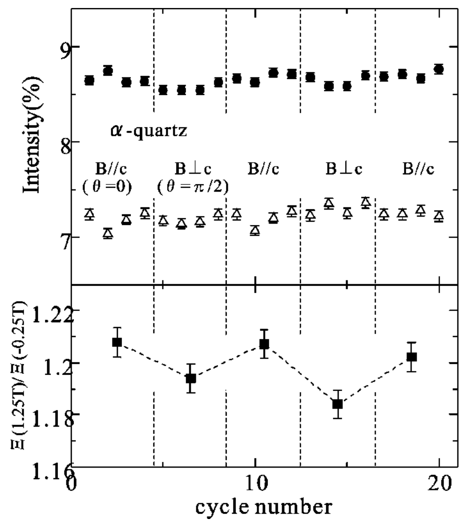


FIG. 2. Top: relative intensity of the positronium peak in $\alpha$ quartz for two different orientations with regard to the magnetic field. (Closed circles: 1.25 T, open triangles: $-0.25$ T.) Bottom: the ratio of the intensity of the positronium peak in the magnetic field of 1.25 T to that in $-0.25$ T. The values are averaged over four scans in each orientation.

peaks around the center reflecting the Bloch nature of the Ps wave function.[9,10] The broad momentum distribution has resulted from Ps pickoff annihilation and positron annihilation without forming Ps. In order to determine the intensity $I_{\mathrm{Ps}}$ of the self-annihilated Ps, we subtracted the broad component assuming it to be a two-dimensional Gaussian distribution.

The results are shown in Fig. 2. The top of this figure shows $I_{\mathrm{Ps}}$ in different magnetic fields; the bottom shows the ratio of $I_{\mathrm{Ps}}$ in a magnetic field of 1.25 T to that in $-0.25$ T. Since Ps atoms are delocalized in quartz, the EFG tensor is axially symmetric ($\eta=0$ and $\Xi$ is $\varphi$ independent), with the principal axis **z**, collinear with the $c$ axis. [In the following, we write $\Xi(B,\theta)$ instead of $\Xi(B,\theta,\varphi)$.] From Eq. (12), the ratio of $I_{\mathrm{Ps}}$ is written as $\Xi(1.25T,\theta=0,$ or $\pi/2)/\Xi(-0.25T,\theta=0,$ or $\pi/2)$, with $\theta=0$ referring to $\mathbf{B}$ being pararell to the $\hat{c}$ axis. To see the angular dependence clearly, the values are averaged over the four scans for each orientation of the crystal. It clearly shows the existence of the quadrupole moment of Ps in $\alpha$ quartz.

We estimate the quadrupole coupling constant $d$ from the anisotropy of $[\Xi(1.25T,\theta)/\Xi(-0.25T,\theta)-1]$. In the weak magnetic field, $\Xi\approx1$ and the field dependence is negligible. Then $[\Xi(1.25T,\theta)/\Xi(-0.25T,\theta)-1]$ can be approximated to $[\Xi(B,\theta)-1]_{B=1.25\ \mathrm{T}}$.

In $\alpha$ quartz $[\Gamma_0(0)/\Gamma_1(0)]y_q^2\ll1$ holds. Then a further approximation

$$\Xi(B,\theta)\approx\frac{\Gamma_0(0)}{\Gamma_1(0)}y_q^2+2Py_q+1, \tag{13}$$

of Eq. (12) is possible. Taking into account the condition $|1-y_q/y|\ll1$, we obtain

$$\Xi(B,\theta=0)\approx\Xi\left(B,\theta=\frac{\pi}{2}\right)+\frac{\partial\Xi}{\partial y_q}\left[y_q(\theta=0)-y_q\left(\theta=\frac{\pi}{2}\right)\right]$$
$$\approx\Xi\left(B,\theta=\frac{\pi}{2}\right)+3y\left(\left[\frac{\Gamma_0(0)}{\Gamma_1(0)}\right]y+P\right)\frac{d}{2\omega\sqrt{1+x^2}}. \tag{14}$$

In view of Eqs. (13) and (14), the anisotropy of $[\Xi(B,\theta)-1]$ is

$$\frac{\Xi(B,\theta=0)-1}{\Xi\left(B,\theta=\frac{\pi}{2}\right)-1}\approx1+\frac{3\left(\left[\frac{\Gamma_0(0)}{\Gamma_1(0)}\right]y+P\right)}{\left[\frac{\Gamma_0(0)}{\Gamma_1(0)}\right]y+2P}\frac{d}{2\omega\sqrt{1+x^2}}. \tag{15}$$

From Fig. 2, we obtain an anisotropy $1.09\pm0.028$. Using the values of $B=1.25$ T, $\kappa=0.31\pm0.02$,[7] $\gamma_{po}=3.66\pm0.12\ \mathrm{ns}^{-1}$,[7] and $P=0.31\pm0.02$,[11] $d$ is determined to be $(3.0\pm0.9)\times10^{-5}$ eV.

Since the hyperfine tensor is written as $A_{ik}=\omega\delta_{ik}+Q_{Ps}V_{ik}$[4], thus the anisotropy $A_{zz}/A_{xx}$ is calculated here as

$$\frac{A_{zz}}{A_{xx}}=\frac{\omega+Q_{\mathrm{Ps}}V_{zz}}{\omega+Q_{\mathrm{Ps}}V_{xx}}=\frac{\omega+d}{\omega-\frac{d}{2}}\approx1.18. \tag{16}$$

The value determined in this study is much smaller than that obtained by Gessmann *et al.* of $A_{zz}/A_{xx}\approx1.93$. Thus our result is not consistent with the large effect reported by Gessmann *et al.* The qualitative agreement is probably accidental due to the poor resolution of the Doppler broadening method they used and their rather complicated way of the data analysis.

The value of $d$ obtained in this work is a few tens of times larger than that estimated by Bondarev and Kuten[4] from the analogy with the Mu quadrupole interaction in $\alpha$-quartz. This is probably because the deformation of the Ps wave function by the crystalline potential in $\alpha$-quartz is much larger than that of the Mu wave function. It is supported by the fact that the contact density of Ps in $\alpha$-quartz is $\sim0.3$, whereas that of Mu is $\sim1$.[6] This value of $d$ yields the ratio $d/\omega_0\approx0.036$, in agreement with the theoretical estimates of Bondarev.[5]

The new experimental technique we employed to detect the effective quadrupole interaction extends potentialities of the positronium spectroscopy of solids. One may experimentally investigate anisotropic hyperfine interaction of the Ps in other materials in the same manner.

In conclusion, we confirmed the existence of the effective quadrupole interaction of Ps induced by the difference in the effective masses of the electron and the positron in material. Anisotropy in the fraction of the self-annihilation of Ps in $\alpha$-quartz was observed by using the 2D-ACAR method. The positronium quadrupole coupling constant $d=Q_{\mathrm{Ps}}V_{zz}$ in $\alpha$-quartz has been determined to be $(3.0\pm0.9)\times10^{-5}$ eV.

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