Positronium Quadrupole Interactions in Crystals *

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It is shown that, due to the hyperfine interaction between the electron and the positron, positronium atoms in a crystal can acquire an effective quadrupole moment and a tensor polarizability. In such a case the effective quadrupolar interaction with intracrystalline fields leads to a quadrupolar splitting of the triplet level of positronium and also to an anisotropy of its magnetic quenching in the presence of an external magnetic field. The possibilities of observating this anisotropy experimentally are discussed.

Key words: Positronium, Quadrupole interaction, Magnetic quenching.

1. Introduction

At the present time the nuclear quadrupole resonance (NQR) covers all the aspects of nuclear quadrupole interactions in gases, liquids and solids. Recently, a new area of NQR investigations has appeared: the study of quadrupole interactions of hydrogen-like (H-like) atoms in a medium. As is known [1], due to the noncentral part of the hyperfine interaction between the electron and the nucleus, all H-like atoms possess quadrupole moments Q in their ground states. For example, Q is equal to 0.7 barn for hydrogen, 0.2 barn for deuterium, and 2.4 barn for muonium. Investigations of the quadrupole interactions of H-like atoms in a medium permit not only to study the intracrystalline fields but also the diffusion mechanism of light interstitials in the crystal lattice and the isotopic effects. The behaviour of H-like atoms (hydrogen, deuterium, muonium) has been well investigated in fluorides, alkali halides and in α-quartz [2, 3]. In particular, these atoms have been shown to occupy the same positions in the α-quartz lattice. In the present paper we would like to dwell on the theoretical consideration of the quadrupole interactions of positronium (Ps), the lightest H-like atom, in noncubic crystals and to discuss the possibilities of their experimental investigation.

Experiment shows that in semiconductors, ionic, and molecular crystals positronium may be in two states: localized or delocalized (of the Bloch wave type) [4]. In crystals Ps atoms experience the action of crystal fields where magnitude and symmetry depend on the Ps state and, in the case of localized positronium, on the rif. Crystal fields lead to an essential change in the structure of hyperfine energy levels of positronium in comparison with the vacuum pattern. In particular, in crystals with noncubic lattice (crystalline quartz, polymers) "anisotropic" positronium [5, 6] may appear. Such positronium atoms are described by an anisotropic hyperfine interaction between the electron and the positron of the type [7]

$$\hat{H} = A_{ik}\, S_{\mathrm{e}}^{i}\, S_{\mathrm{p}}^{k}\,, \tag{1}$$

where A_{ik} is the matrix of the hyperfine interaction, and $\boldsymbol{S}_{\mathrm{e}}, \boldsymbol{S}_{\mathrm{p}}$ are the spin operators of the electron and the positron, respectively. Due to the action of the crystal field the triplet level of "anisotropic" positronium splits even in the absence of an external magnetic field. Below we shall show that the physical cause of such an anisotropy of the hyperfine interaction of Ps in a crystal is the existence of the effective quadrupole moment and the tensor polarizability in its ground state.

2. Effective Quadrupole Interaction of Positronium in a Crystal

Let us consider the hyperfine structure of Ps energy levels (ground state) in a crystal. It is convenient to do this by the spin Hamiltonian method. For a Ps atom

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the Hamiltonian of the hyperfine interaction between the electron and the positron takes the form [8]

$$\hat{H} = 2\pi\,\mu_e\,\mu_p\left(3+\frac{7}{3}(\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_p)\right)\delta(\boldsymbol{r}) + \frac{\mu_e\,\mu_p}{r^3}\left(3(\boldsymbol{\sigma}_e\cdot\boldsymbol{n})(\boldsymbol{\sigma}_p\cdot\boldsymbol{n})-(\boldsymbol{\sigma}_e\cdot\boldsymbol{\sigma}_p)\right), \tag{2}$$

where μ_e and μ_p are the magnetic moments of the electron and the positron, respectively, $\boldsymbol{\sigma}_e$ and $\boldsymbol{\sigma}_p$ are the Pauli matrices, and $\boldsymbol{n}$ is the unit vector in the $\boldsymbol{r}$ direction. In crystals the radius of excitonic Ps-like states is small [9, 10]. For this reason one should change the masses of the electron and the positron in (2) by their effective masses m_e and m_p from the theory of small radius positronium exciton [10].

It is easy to see that the noncentral part of the interaction (2) provides a small D-wave admixture (orbital moment $l=2$) to the triplet S-state of Ps (total angular moment $F=1$). As a result, the exact triplet ground state of positronium is not spherically symmetric and takes the form [1]

$$\psi_0^{F=1,m_F}(\boldsymbol{r}) = R_S(r)\,(Y_0(\boldsymbol{n})\otimes\chi_1)^{F=1,m_F} + R_D(r)\,(Y_2(\boldsymbol{n})\otimes\chi_1)^{F=1,m_F}, \tag{3}$$

where $R_S(r)$, $R_D(r)$, $Y_0(\boldsymbol{n})$, $Y_2(\boldsymbol{n})$, χ_1 are the radial parts, the spherical harmonics of rank 0 and 2 and the spin part (total spin S is equal to 1) of the Ps wave function, respectively.

We see that the quadrupole moment positronium, which is defined (in the centre-of-mass system) as [7, 11]

$$\begin{aligned} Q_{Ps} &= \frac{m_e-m_p}{m_e+m_p}\langle\psi_0^{F=1,m_F=1}(\boldsymbol{r})\,|3z^2-r^2|\,\psi_0^{F=1,m_F=1}(\boldsymbol{r})\rangle \\ &\approx 2\,\frac{m_e-m_p}{m_e+m_p}\langle R_S(r)\,(Y_0(\boldsymbol{n})\otimes\chi_1)^{F=1,m_F=1}\,|3z^2-r^2| \\ &\quad\cdot R_D(r)\,(Y_2(\boldsymbol{n})\otimes\chi_1)^{F=1,m_F=1}\rangle, \end{aligned} \tag{4}$$

is not equal to zero if the effective masses of the electron and the positron are not the same. Just this situation prevails in a crystals because of the different character of the electron and the positron interaction with the surrounding electrons of the medium. It is interesting to note that due to the equality of the electron and the positron masses and also due to the execution of the CP- and T-invariance laws in the system "particle-antiparticle", the free Ps in vacuum is a completely electroneutral system and cannot have any multipole moments [8] (excluding the anapole moment, which arises because of parity nonconservation [12]). In a crystal the effective masses of the electron and the positron are, generally speaking, not the same. As a consequence, positronium atoms in a crystal acquire a new fundamental characteristic – quadrupole moment [13].

But the presence of the quadrupole moment is not the only manifestation of the hyperfine interaction between the electron and the positron in Ps. It also manifests itself in the presence of the atomic tensor polarizability (see the analogous phenomenon for hydrogen in [14]), which is proportional to the dipole moment of Ps induced by the intracrystalline electric fields (we consider the splitting of the level only, but not the shift of the level caused by the action of the crystal field [15, 16]). Thus the interaction of the Ps atom with a crystal can be described by the interaction of its induced dipole moment with the intracrystalline electric field (the Stark effect on the hyperfine structure) and also of its quadrupole moment with the electric field gradient (EFG).

The corresponding spin Hamiltonian takes the form [7, 11, 17] ($\hbar=1$)

$$\hat{H} = \omega\,\boldsymbol{S}_e\cdot\boldsymbol{S}_p + \frac{Q_{Ps}}{4}\,Q_{ik}\,\varphi_{ik} - \frac{\alpha_t}{2}\,Q_{ik}\,\varepsilon_i\,\varepsilon_k - \frac{1}{2}\,i\Gamma, \tag{5}$$

where ω is the frequency of the hyperfine splitting of Ps in the crystal (in vacuum this value is equal to $\omega_0 = 8.2\cdot 10^{-4}$ eV [18]), $\varphi_{ik} = \partial^2\varphi(0)/\partial x_i\,\partial x_k$ is the EFG tensor in the centre of mass of the Ps atom ($\varphi(0)$ is the corresponding potential of the environment), $\varepsilon_i = \partial\varphi(0)/\partial x_i$ is the intracrystalline electric field, $Q_{ik} = F_i F_k + F_k F_i - \frac{2}{3}F(F+1)\,\delta_{ik}$, $\boldsymbol{F} = \boldsymbol{S}_e + \boldsymbol{S}_p$ is the total angular moment of Ps, Q_{Ps} and α_t are the quadrupole moment and the tensor polarizability, respectively (in vacuum $\alpha_t = -0.74\cdot 10^{-28}$ cm^3 – one can get this value from the tensor polarizability of hydrogen [14]), and Γ is the decay part of the Hamiltonian.

The last two terms in (5) may be reduced to the effective quadrupole interaction $(Q_{Ps}/4)\,Q_{ik}\,\tilde{\varphi}_{ik}$, where $\tilde{\varphi}_{ik} = \varphi_{ik} - (2\alpha_t/Q_{Ps})\,\varepsilon_i\,\varepsilon_k$ is the tensor of the electric fields and their gradients (EFTG). It consists of two parts: the EFG tensor and the part related to the intracrystalline electric fields. In a delocalized positronium state the contribution of electric fields to the EFTG tensor is apparently defined by the symmetry of the crystal. In particular, this contribution evidently equals zero in crystals with the inversion centre. In a localized positronium state it is equal to zero (or very small), since it is natural to suppose that the Ps atom is localized in the minimum of potential of its environment.

Using the anticommutation relations for spin $-\frac{1}{2}$ operators.

$$\{S_i, S_k\}=\tfrac{1}{2}\delta_{ik}$$

(δ_{ik} is the Kronecker δ, $i, k=x, y, z$), one can transform (5) into

$$\hat{H}=(\omega\,\delta_{ik}+Q_{\mathrm{Ps}}\,\tilde{\varphi}_{ik})\,S_{\mathrm{e}}^{i}\,S_{\mathrm{p}}^{k}-\tfrac{1}{2}\,i\,\Gamma\,. \tag{6}$$

Comparing (6) and (1) we obtain the connection between the phenomenological constants A_{ik} and the characteristics of the effective quadrupole interaction of Ps in a crystal:

$$A_{ik}=\omega\,\delta_{ik}+Q_{\mathrm{Ps}}\,\tilde{\varphi}_{ik}\,. \tag{7}$$

Thus one can say that the physical cause of the anisotropy of the hyperfine interaction of Ps in a crystal is the existence of the effective quadrupole moment and the tensor polarizability in the ground state of positronium.

In the presence of an external magnetic field $\boldsymbol{B}$ we have to add to (5) the terms describing the interaction of Ps with it:

$$\hat{H}=\omega\,\boldsymbol{S}_{\mathrm{e}}\cdot\boldsymbol{S}_{\mathrm{p}}-\boldsymbol{\mu}_{\mathrm{e}}\cdot\boldsymbol{B}-\boldsymbol{\mu}_{\mathrm{p}}\cdot\boldsymbol{B}+\frac{Q_{\mathrm{Ps}}}{4}\,Q_{ik}\,\tilde{\varphi}_{ik}-\tfrac{1}{2}\,i\,\Gamma\,. \tag{8}$$

In [19] a detailed analysis of the energy levels of the spin Hamiltonian (8) was made for the case when the magnetic field is parallel to the principle axis of the $\tilde{\varphi}_{ik}$-tensor, Z. Unfortunately exact analytical solution of the eigenvalue problem given by (8) is impossible for an arbitrary direction of $\boldsymbol{B}$ with respect to the system of principle axes of the EFTG tensor. Therefore we shall consider the interaction of Ps with a crystal field (the effective quadrupole interaction) by means of perturbation theory. Then the levels and the eigenstates of (8) are easily found and take the form (in the system of principle axes of the EFTG tensor) ($\hbar=1$)

$$\begin{aligned}
E_{0,1}&=-\frac{\omega}{4}\mp\frac{\omega}{2}\sqrt{1+x^{2}}-\frac{\tilde{d}}{4}\,C_{1,0}^{2}\,\phi(\vartheta,\varphi)\,,\\
E_{2,3}&=\frac{\omega}{4}\pm(\mu_{\mathrm{e}}+\mu_{\mathrm{p}})\,B+\frac{\tilde{d}}{8}\,\phi(\vartheta,\varphi)\,,\\
\gamma_{0,1}&=\frac{\gamma_{\mathrm{s,t}}+y^{2}(\vartheta,\varphi)\,\gamma_{\mathrm{t,s}}}{1+y^{2}(\vartheta,\varphi)}\,,\quad \gamma_{2}=\gamma_{3}=\gamma_{\mathrm{t}}\,,
\end{aligned} \tag{9}$$

where

$$C_{0,1}=\sqrt{(1/2)\,(1\pm1)/\sqrt{1+x^{2}}}\,,$$

$$y^{2}(\vartheta,\varphi)=y_{0}^{2}\,(1+\tilde{d}\,\phi(\vartheta,\varphi)/2\,\omega\sqrt{1+x^{2}}),$$

will

$$y_{0}=(\sqrt{1+x^{2}}-1)/x,\; x=2(\mu_{\mathrm{e}}-\mu_{\mathrm{p}})\,B/\omega\,.$$

γ_{s} and γ_{t} are the decay widths of singlet and triplet levels of Ps in matter, respectively. (Due to the distortion of Ps wave function in matter, they, generally speaking, differ from the vacuum decay widths $\gamma_{\mathrm{s}}^{0}=8\cdot10^{9}\ \mathrm{s}^{-1}$ and $\gamma_{\mathrm{t}}^{0}=7.14\cdot10^{6}\ \mathrm{s}^{-1}$.) Furthermore

$$\phi(\vartheta,\varphi)=\left(1+\frac{2\,\alpha_{\mathrm{t}}\,\varepsilon^{2}}{3\,d}\right)(3\cos^{2}\vartheta-1)+\tilde{\eta}\,\sin^{2}\vartheta\,\cos2\,\varphi\,,$$

where ϑ and φ are the polar and lateral angles characterizing the tilt of $\boldsymbol{B}$ with respect to the principal axis of the EFTG tensor. $\tilde{d}=Q_{\mathrm{Ps}}\,\tilde{\varphi}_{zz}$, $\tilde{\eta}=|(\tilde{\varphi}_{xx}-\tilde{\varphi}_{yy})/\tilde{\varphi}_{zz}|$ are the effective quadrupole constant of positronium and the asymmetry parameter of the EFTG tensor [11].

We see from (9) that in the presence of an external magnetic field the effective quadrupole interaction leads to an anisotropic splitting of the hyperfine levels of Ps. This anisotropy is determined by the angle between the $\boldsymbol{B}$ direction and the principle axis of the EFTG tensor. The anisotropic splitting of the hyperfine levels of Ps leads to oscillations in the count rate of the quanta of 3γ-decay with frequencies that depend on the angle between $\boldsymbol{B}$ and Z. (This phenomenon was analyzed in [19].)

It also follows from (9) that in the presence of an external magnetic field the decay widths of Ps are anisotropic and depend on the magnetic field direction with respect to the system of principle axes of the EFTG tensor. As a consequence, in a crystal we have the phenomenon of anisotropic magnetic quenching of positronium. In particular, at $\phi(\vartheta,\varphi)=0$, i.e. if

$$\sin^{2}\vartheta=\frac{2(1+2\,\alpha_{\mathrm{t}}\,\varepsilon^{2}/3\,\tilde{d})}{3(1+2\,\alpha_{\mathrm{t}}\,\varepsilon^{2}/3\,\tilde{d})-\tilde{\eta}\,\cos2\,\varphi} \tag{10}$$

holds (Dean cone), the effective quadrupole interaction does not contribute to the hence magnetic quenching and the magnetic quenching of Ps must here be isotropic. At $\tilde{\eta}=0$ (EFTG tensor is axially symmetric) the semivertex angle of the cone (10) is 54°44′. If $\phi(\vartheta,\varphi)\neq0$ the character of the magnetic quenching of Ps changes due to the effective quadrupole interaction. The processes of quenching differ maximally when the magnetic field is parallel or perpendicular to Z, the principle axis of the EFTG tensor.

3. Conclusion

We have considered the action of a crystal field on the Ps atom by means of perturbation theory. However, such a consideration is not always correct, generally speaking. For instance, experience shows that the wave function of the "anisotropic" Ps in polymers is strongly distorted in comparison with the vacuum one [6] (the strong crystal field). Consequently, the correct theoretical analysis of the hyperfine structure of Ps levels is only possible by means of the quantum chemical methods (one should note, however, that the anisotropic splitting of Ps levels and the anisotropy of the magnetic quenching of Ps take place in this case as before). Nevertheless, an experiment shows sometimes that the Ps wave function in a crystal is weakly distorted (in a weak crystal field; for example in crystalline quartz [20, 21]. In such a case the consideration of the interaction of positronium with a lattice by means of perturbation theory is quite correct.

For quartz we can even get estimations of the effects described above. The observation of the oscillations in the count rate of the quanta of 3γ-decay is strongly embarrassed in quartz because of the large pick-off annihilation rate of positronium ($\gamma_p = 0.88 \cdot 10^9\ \mathrm{s}^{-1}$ [20]). However the effective quadrupole interaction of Ps in quartz can be experimentally investigated by means of the observation of the anisotropy of the magnetic quenching. In a weak external magnetic field ($x^2, y^2(\vartheta, \varphi) \ll 1$) the magnetic quenching can be characterized by the magnetic quenching parameter [22]. For quartz it takes the form

$$Q(\vartheta, \varphi) = y^2(\vartheta, \varphi)\,\frac{\gamma_s}{\gamma_t}\,,$$

where $\gamma_s \approx \gamma_s^0 + \gamma_p$, $\gamma_t \approx \gamma_t^0 + \gamma_p$ [9] with γ_p the pick-off annihilation rate of Ps in quartz. The relative decrease of the probability of 3γ-annihilation (W), the enhancement of the narrow component of the ACAR-curve (E), and the suppression of the long-living component in the time spectrum (R), measured in the experiments on the magnetic quenching, are defined by this parameter [22] as

$$W(B) = \frac{2}{3} + \frac{1}{3(1 + Q(\vartheta, \varphi))}\,, \quad E(B) = \frac{Q(\vartheta, \varphi)}{1 + Q(\vartheta, \varphi)}\,,$$

$$R(B) = \tfrac{1}{3}\left(2 + \mathrm{e}^{-Q(\vartheta, \varphi)}\right).$$

In our case, taking into account the conditions $x^2 \ll 1$ and $\gamma_t^0 \ll \gamma_p \ll \gamma_s^0$, we obtain

$$Q(\vartheta, \varphi) \approx \frac{x^2 \gamma_s^0}{4\gamma_p}\left(1 + \frac{\tilde{d}\,\phi(\vartheta, \varphi)}{2\omega}\right) B^2$$

$$\approx 1.71 \cdot 10^{-3}\left(1 + \frac{d(3\cos^2\vartheta - 1)}{2\omega}\right) B^2\,,$$

where the magnetic field is measured in kG (for the sake of simplicity, we put $\tilde{\eta} = 0$ and took into account that the Stark splitting of Ps levels is considerably smaller than the quadrupole splitting, i.e. $\tilde{d} \approx d$). Consequently, the difference between the magnitudes of the magnetic quenching parameter in a parallel ($\vartheta = 0$) and perpendicular ($\vartheta = \pi/2$) magnetic field is

$$\Delta Q = Q(\vartheta = 0) - Q\left(\vartheta = \frac{\pi}{2}\right) \approx 2.57 \cdot 10^{-3}\,\frac{d}{\omega}\,B^2\,.$$

The magnitude of the parameter d/ω for Ps in quartz can be estimated using the analogous data for muonium [2]. According to [2], for Mu in quartz $d^{\mathrm{Mu}}/\omega^{\mathrm{Mu}} \approx 2.08 \cdot 10^{-3}$. For Ps the following takes place: the D-wave admixture forming the quadrupole moment of Ps in its ground state (see (3), (4)) is of the order of $\mu_e\,\mu_p$, whereas for Mu it is of the order of $\mu_e\,\mu_\mu$. Taking into account that $m_\mu \approx 206.77\,m_e$, we obtain $Q_{\mathrm{Ps}} \approx 206.77\ Q_{\mathrm{Mu}}$ (we suppose that the mass factor in Q_{Ps} (4) is of the order of 1). Analogously one can obtain $Q_{\mathrm{Ps}} \approx 659.50\ Q_{\mathrm{H}}$. If we roughly assume now that in quartz the gradient of the intracrystalline electric field is the same for Mu and for Ps, we obtain $d/\omega \approx 4.73\ d^{\mathrm{Mu}}/\omega^{\mathrm{Mu}} \approx 9.84 \cdot 10^{-3}$ (we used the vacuum value for the frequency of the hyperfine splitting of Ps in quartz). As a result, the difference between the magnitudes of the magnetic quenching parameter of Ps in parallel and perpendicular field is equal to

$$\Delta Q = 2.53 \cdot 10^{-5}\,B^2\,.$$

For $B \approx 20$ kGs we have $\Delta Q \approx 1\%$. In such a case it is easy to see that the differences of the experimental parameters W, E, and R for parallel and perpendicular magnetic field are

$$\Delta W \approx 0.3\%\,, \quad \Delta E \approx 1\%, \quad \Delta R \approx 0.3\%\,,$$

respectively.

Unfortunately, due to the weak distortion of the wave function, it is difficult to expect a strong anisotropy of the magnetic quenching of Ps in quartz. But, for example, in polymers [6] the distortion of the Ps wave function is very large and this phenomenon should manifest itself much more strongly.

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