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Effect of host medium on the L/K ratio in ^{7}Be electron capture

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We discuss in the framework of linear muffin-tin orbital method and Bahcall's calculations how the ratio of L- to K-shell electron capture rates (L/K ratio) in ^{7}Be would be affected by the host medium in which ^{7}Be is implanted. Our calculations show that the recently observed discrepancy between the measured and observed L/K ratio in ^{7}Be could be understood quantitatively as a result of in-medium effects distorting the L-shell electron orbital of ^{7}Be.

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The study of electron capture by ^{7}Be nucleus is a topic of current interest and has contributed to many fields such as the development of solar model, test of weak interaction theory, etc. ^{7}Be is the lightest radioactive nucleus that decays by electron capture. It would be of special interest to understand L/K capture ratio in ^{7}Be, because at low atomic number the correction factor (to L/K ratio) for exchange and overlap effects becomes very large. So such studies will test the basic theories of exchange and overlap corrections at low atomic number.

Recently Voytas *et al.* [1] implanted ^{7}Be in mercury telluride (HgTe) and measured the ratio of L- to K-shell electron capture rates of ^{7}Be using a cryogenic microcalorimeter. They found the L/K capture ratio to be 0.040 ± 0.006.

The ratio [2] of L to K capture determined solely from Hartree's [3] calculated $1s$ and $2s$ radial wave functions (having two full $1s$ and $2s$ electrons) of ^{7}Be atom is $=0.0331$. However, exchange and overlap corrections to L/K ratio are very important for low atomic number nuclei and cannot be neglected. At present, there are two basic theoretical approaches [4], one due to Bahcall and another due to Vatai, for calculating exchange and overlap corrected L/K ratio for electron capturing nuclei. Bahcall's technique yields $L/K=0.09$ for a free ^{7}Be atom having two full $1s$ and $2s$ electrons. Vatai's approach [4] neglects some contributions involving shakeup or shakeoff and uses perturbation theory to calculate the exchange integrals. Vatai's technique gives $L/K=0.11$ for a free ^{7}Be with two full $1s$ and $2s$ electrons. So the experimentally measured value [1] of L/K ratio is less than half of the predicted theoretical values [4].

Voytas *et al.* [1] speculated that the distortion of L-shell electron orbitals of ^{7}Be due to in-medium effects might be responsible for such discrepancy. However, it is very important to understand this discrepancy quantitatively and find out if the discrepancy is indeed due to the effect of host medium. It is well known [5–7] from the earlier studies on the half-life of ^{7}Be in different media that the half-life of ^{7}Be changes due to the distortion of its $2s$ (L-shell) electron orbitals by the host media. However, such change of half-life of ^{7}Be has so far been found to be less than 1% [5–7].

Such in-medium effect is expected to be more dramatic for L- to K-shell electron capture ratio (L/K ratio) in ^{7}Be, because only $2s$ electrons (L-shell electrons) of ^{7}Be are seriously affected by such effects whereas $1s$ electrons (K-shell electrons) of ^{7}Be are hardly influenced by in-medium effects. As a result, the ratio of L- to K-shell electron capture rates in ^{7}Be nucleus is expected to be rather sensitive to the host medium where ^{7}Be is implanted.

In this paper, we show that the L/K ratio in ^{7}Be can indeed drop significantly from its ideal theoretical value, because on the average, ^{7}Be atoms lose a very significant fraction of their $2s$ electrons as a result of implantation of ^{7}Be in a medium. Let us first discuss this problem qualitatively in terms of electron affinity and lattice dimensions.

The electron affinity of beryllium is -0.19 eV [8]. The negative value of electron affinity for beryllium means that there is no bound state of an extra electron to the ground state of the beryllium atom. The average number of $2s$ electrons that ^{7}Be loses, when implanted in a medium, depends on the electron affinity and lattice structure of the host medium. A beryllium atom implanted in a medium having high electron affinity will lose a larger fraction of its $2s$ electrons compared to a beryllium atom implanted in a medium of low electron affinity. The dimensions and structure of the host lattice also matter since it is important how close to a host atom ^{7}Be sits. Usually a ^{7}Be atom is expected to lose more $2s$ electrons if it sits closer to a host atom. Although mercury telluride does not have much electron affinity, but even then implanted ^{7}Be atoms in mercury telluride might lose a considerable fraction of their $2s$ electrons (L-shell electrons) because of the presence of nearby host atoms and slightly negative electron affinity of beryllium. Since K-shell electrons ($1s$ electrons) of ^{7}Be remain essentially unaffected by such in-medium effects, hence significant reduction of L/K electron cature ratio in ^{7}Be is possible in the case of implantation of ^{7}Be in mercury telluride.

We shall now use linear muffin-tin orbital (LMTO) method [6,9] to determine the average number of $2s$ electrons in a ^{7}Be atom when it is implanted in a medium. In the tight binding linear muffin-tin orbital (TBLMTO) method [9], the interatomic potential is assumed to be of muffin-tin type and written as

$$V_{MT}(r)=V_i(r_i)+\sum_R V_R(r_R)\equiv V_0+\sum_R v_R(r_R), \quad (1)$$

where $V_R(r_R)$ and $v_R(r_R) \equiv V_R(r_R) - V_0$ are spherically symmetric inside a sphere of radius s_R centered at R and vanish outside. $V_i(r_i)$ takes the constant value V_0 (the muffin-tin zero) in the interstitial region and vanishes outside. A beryllium atom is put in the interstitial region and a spherically symmetric potential is considered centered around this atom. This spherical potential also vanishes outside a certain radius. Schrödinger's equation was solved for this problem assuming periodic boundary condition. Atomic muffin-tin orbitals have been considered spherical and no deformation due to the overlap of two nearby muffin-tin orbitals has been considered. For a given position of the implanted beryllium atom and the assumption of spherical potential, LMTO method performs a first-principles calculation and there is no adjustable free parameter in the calculation. Let Ψ_{total} be the complete electronic wave function and Ψ_{Be2s} be beryllium $2s$ state wave function. We calculate the square of the overlap of Ψ_{total} with Ψ_{Be2s}, i.e $|\langle\Psi_{total}|\Psi_{Be2s}\rangle|^2$, which represents the average number of $2s$ electrons in a beryllium atom when it is implanted in the medium. The average number of beryllium $2s$ electrons is expected to depend on the position of beryllium atom in the interstitial region.

We apply this method to calculate average number of $2s$ electrons in a ^{7}Be atom when it is implanted in HgTe. The calculations have been done assuming that the atoms are in their ground states. Since Voytas *et al.* [1] have done measurements at 60 mK temperature, so we can certainly neglect any thermal excitation of the atoms. The structure, lattice dimensions, and space group of mercury telluride are inputs to the code. In addition, the atomic structures of Hg, Te, and Be are also needed. It is well known that the structure of mercury telluride comprises [8] two face-centered cubic lattices of Hg and Te displaced from each other by one-quarter of a body diagonal. The lattice parameter of mercury telluride at room temperature is 6.4623 Å [8] and its space group is F-43m. Although ideally, we should have used the value of lattice parameter at 60 mK temperature (where measurements have been done), but only the value of lattice parameter at room temperature (300 K) is available. So that number was used for our calculations. The coefficient of thermal linear expansion of HgTe is also not known. In order to see the effect of temperature, we have made a rough estimate of the lattice parameter of HgTe at 60 mK temperature by doing a linear extrapolation from room temperature using the known coefficient of thermal linear expansion of zinc telluride. The reduction of the lattice parameter has been found to be 0.15%. Although this is an underestimation, but we found that even a 0.5% reduction in lattice dimension has insignificant effect ($\approx$0.1%) on our final results. So we do not consider any reduction of lattice dimension at low temperature in our calculations.

We assume that the implanted ^{7}Be will go to either octahedral (center of the lattice) or tetrahedral positions and not more than one ^{7}Be is implanted in a particular lattice. When ^{7}Be is in an octahedral site, then the volume overlap of ^{7}Be atom with its nearest host atom has been found to be 14%. In the case of implantation in a tetrahedral site also, the overlap has been found to be about the same. It is very unlikely that ^{7}Be would go anywhere other than octahedral or tetrahedral sites, because of the Coulomb repulsion between a ^{7}Be atom and its nearest host atom due to their significant volume overlap. The ratio of overlap volume between the nearest atoms to the total volume enters into linear muffin-tin orbital method calculation as a perturbation term and hence it is not reasonable to consider very large volume overlap.

Our calculations show that when ^{7}Be is in an octahedral site of mercury telluride crystal, then the square of the overlap of Ψ_{total} with Ψ_{Be2s}, i.e $|\langle\Psi_{total}|\Psi_{Be2s}\rangle|^2 = 1.047$. The quantity $|\langle\Psi_{total}|\Psi_{Be2s}\rangle|^2$ represents the average number of $2s$ electrons in a ^{7}Be atom. When ^{7}Be is in a tetrahedral position of mercury telluride crystal, then we find $|\langle\Psi_{total}|\Psi_{Be2s}\rangle|^2 = 1.209$. Since the number of tetrahedral sites are twice that of octahedral sites, so in the case of random implantation, ^{7}Be is twice more likely to go to a tetrahedral site. So taking weighted average, we find that the average number of $2s$ electrons in ^{7}Be would be $=1.155$ instead of 2.0 as in the case of a free ^{7}Be atom. Since fewer $2s$ electrons are available, so this will lead to a reduction of $2s$ (L-shell) electron capture rate by a factor of (1.155/2.0) $=0.577$ compared to that in a completely free ^{7}Be atom with two full $1s$ and $2s$ electrons.

K-shell electron capture rate should essentially remain unchanged. In principle, the removal of electrons from $2s$ orbital should increase the electron capture rate of $1s$ electrons by a very small amount, because the screening of the nuclear charge by $2s$ electrons decreases. However, for the same reason, L-shell decay rate should also increase slightly, since there will be lesser amount of mutual screening between two $2s$ electrons. So L/K ratio is not expected to change much because of such change in screening correction. Moreover, these effects are extremely tiny. Bahcall [2] discussed the question of change of K-shell decay rate of ^{7}Be due to the removal of electrons from $2s$ orbital and found that the value of $1s$ electron wave function at the nucleus, i.e, $\Psi_{Be1s}(r=0)$ is essentially the same (within a few tenths of a percent) for both neutral Be and Be^{++} (both $2s$ electrons removed). This happens because when an electron is at the nucleus or very close to it, then it experiences essentially unscreened total nuclear charge. We have neglected such small effects in our L/K calculations.

We think that the effect on the exchange term [10] (for L/K ratio) due to the removal of one $2s$ electron is also very small. Considering the change of normalization constant of the wave function of $2s$ orbital due to the removal of one electron, the exchange correction factor [10] decreases by about 2% only. However, on the other hand, the change in screening effect (because of removal of electron from $2s$ orbital) should increase the exchange correction factor very slightly. So the overall effect on L/K ratio would be very small and this has not been considered. The effect of imperfect atomic overlap largely cancels out [10] from the electron capture ratio and so the effect of removal of electron from $2s$ orbital on imperfect atomic overlap has not been considered.

So in the zeroth order, L/K ratio should be corrected for in-medium effect by multiplying L/K ratio of neutral ^{7}Be atom by the ratio of the available average number (1.155) of

TABLE I. Calculated L/K ratio of ^{7}Be in different media.

^{7}Be implanted in	Correction factor for in-medium effect	In-medium effect corrected (L/K) ratio	
		Bahcall's calculation	Vatai's calculation
HgTe[a]	0.577	0.0519	0.0635
Al_2O_3	0.432	0.0389	0.0475
^{9}Be	0.4155	0.0374	0.0457
LiF	0.3695	0.0333	0.0406
Al	0.344	0.0310	0.0378
Ta	0.2986	0.0269	0.0328
Au	0.208	0.0187	0.0229

[a]Experimental value of L/K ratio in HgTe is 0.040 ± 0.006.

$2s$ electrons in an implanted Be atom to the number (2.0) of $2s$ electrons in a free and neutral Be atom, i.e., $(1.155/2.0) = 0.577$. So when ^{7}Be is implanted in mercury telluride, then Bahcall's result [4] for $L/K = 0.09$ should be changed to 0.0519 and Vatai's result [4] for $L/K = 0.11$ should be changed to 0.0635. Comparing with Voytas *et al.*'s [1] measured value of $L/K = 0.040 \pm 0.006$, we find reasonable agreement (within two standard deviations) with Bahcall's calculation [4] after multiplying Bahcall's result by the in-medium correction factor of 0.577. Even after applying such in-medium correction factor, Vatai's calculation still seems to be somewhat off from the experimental value. However, Vatai's calculation [4] neglects some contributions involving shakeup or shakeoff effects, uses perturbation theory to calculate exchange integrals, and does rough estimates of overlap corrections. In Bahcall's approach also [4], one uses closure approximation without correction for occupied states. This might be a problem for low-Z nuclei.

Earlier, we performed [6,11] linear muffin-tin orbital method calculations to determine the effect on the L-shell electronic orbital of ^{7}Be when it is implanted in Al_2O_3, LiF, Au, Al, Ta, and natural beryllium. From those calculations, we can determine in-medium effect corrected L/K ratio in ^{7}Be when ^{7}Be is implanted in those media. In Table I, we tabulate theoretical L/K ratios of ^{7}Be in different media after doing in-medium correction (using TBLMTO code) to Bahcall and Vatai's results [4]. At present, experimental L/K ratio [1] is available only for the case of implantation of ^{7}Be in HgTe.

Let us finally discuss the inherent uncertainties in our LMTO method of calculation [9]. The uncertainties in the LMTO method [9] are very small. The most important uncertainty of our method comes from the positioning of Be atom in the host lattice. As one moves a Be atom closer to a host atom, more electrons are removed from the $2s$ orbital of Be atom, which is because of the electron affinity of the host atom. However, for HgTe lattice, there is essentially no space to move around Be atom from its octahedral or tetrahedral sites. In addition, the electron affinities of Hg and Te are very small [8]. Hence our result for HgTe should be robust. In the case of Au lattice calculations, there is a lot of space available around the octahedral site and the electron affinity of Au is large (2.3 eV) [8]. So in that case [6], we moved Be atom to 14 different positions around the octahedral site, determined the number of $2s$ orbital electrons in each case, and then took average.

Other assumptions of LMTO method [9] are the use of spherical potential, the treatment of combined correction factor due to interstitial potential as a perturbation, and the treatment of overlap volume in perturbation. All these uncertainties are generally considered to be very small [9] and neglected.

In conclusion, our calculation shows that ^{7}Be loses a significant fraction of its $2s$ electrons even when it is implanted in a medium such as mercury telluride having essentially no electron affinity. We have been able to understand quantitatively the discrepancy between the measured L/K value [1] of ^{7}Be and theoretical calculations [4]. After doing in-medium corrections, we find that Bahcall's calculation agrees reasonably well with experimental result but Vatai's calculation seems to be off. The success of our linear muffin-tin method calculations to understand these effects puts our earlier conclusion [6] regarding the reduction of predicted ^{8}B solar neutrino flux by $\approx 2\%$ on a more solid basis.

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