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Positron Cooling and Annihilation in Noble Gases

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Positron cooling and annihilation in room temperature noble gases is simulated using accurate scattering and annihilation cross sections calculated with many-body theory, enabling the first simultaneous probing of the energy dependence of the scattering and annihilation cross sections. A strikingly small fraction of positrons is shown to survive to thermalization: ~0.1 in He, ~0 in Ne, ~0.15 in Ar, ~0.05 in Kr, and ~0.01 in Xe. For Xe, the time-varying annihilation rate $\dot{Z}_{\text{eff}}(\tau)$ is shown to be highly sensitive to the depletion of the momentum distribution due to annihilation, conclusively explaining the long-standing discrepancy between gas-cell and trap-based measurements. Overall, the use of the accurate atomic data gives $\dot{Z}_{\text{eff}}(\tau)$ in close agreement with experiment for all noble gases except Ne, the experiment for which is proffered to have suffered from incomplete knowledge of the fraction of positrons surviving to thermalization and/or the presence of impurities.

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The observation of lifetime spectra for positrons annihilating in gases was one of the first sources of information on positron-atom and molecule interactions [1–6]. In particular, measurements of the time-varying normalized annihilation rate $Z_{\text{eff}}(t)$ during positron thermalization provided information on the energy dependence of the scattering and annihilation cross sections. Understanding the dynamics of positron cooling, including the fraction of positrons surviving to thermalization, is critical for the accurate interpretation of the positron lifetime experiments. Incomplete thermalization was suspected to be responsible for the lack of consensus among the $Z_{\text{eff}}$ data in Xe [7], while modeling of $Z_{\text{eff}}(t)$ [8,9] revealed deficiencies in the theoretical data for neon and the heavier noble-gas atoms. Understanding positron kinetics is also crucial for the development of efficient positron cooling in traps and accumulators [10] and for a cryogenically cooled, ultra-high-energy-resolution, trap-based positron beam [11,12].

Despite the importance of long-standing positron-cooling measurements [5,6], there has been a paucity of theoretical studies of positron cooling in gases. Previous studies have mainly employed the diffusion or Fokker-Planck (FP) equation [8,9,13–16]. They used semiempirical or model cross sections, e.g., calculated in the polarized-orbital approximation [17–21], yielding limited success in describing experiments.

Many-body theory (MBT) has provided an accurate and essentially complete description of low-energy positron interactions with noble-gas atoms, taking full account, ab initio, of the strong positron-atom and electron-positron correlations, including virtual-positronium formation [22–25]. Recently, it yielded excellent agreement between theory and experiment for the scattering cross sections, annihilation rates [24,25], and γ-ray spectra [25,26].

Below the positronium (Ps)-formation threshold, energy loss in atomic gases proceeds via momentum transfer in elastic collisions. The process of positron thermalization in a Maxwellian gas of temperature $T$ is governed by the mean-squared change in momentum per unit time $\langle \Delta k^2 / \Delta t \rangle = 2B(k)$, where $B(k) \equiv k\sigma_t(k)M/kM$ [33], $k$ and $m$ are the positron momentum and mass, respectively, $M$ is the mass of the gas atom, and $\sigma_t(k)$ is the positron-atom momentum-transfer cross section. It is calculated as $\sigma_t = 4\pi k^2 \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_l - \delta_{l+1})$ [34], where $\delta_l(k)$ is the scattering phase shift for a positron of angular momentum $l$. Figure 1(a) shows $B(k)$ for He to Xe, calculated using phase shifts for $l = 0, 1,$ and 2 from

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is expected to be negligible (is unimportant until the positrons have cooled close to the annihilation rate is expected to be $\sim 10\%$, although the contribution of positron cooling below the Ps-formation threshold: the elastic-scattering cross section, which is determined from the integral of the differential cross section $q = d\sigma_{el}/d\Omega = |f(\theta)|^2$, where $f(\theta)$ is the scattering amplitude for scattering angle $\theta$, which is calculated using MBT [see Eq. (31) of Ref. [24]]. If a collision occurs, a second random number $r_2 = U[0, 1]$ is drawn. If $r_3 < \sigma_{el}/\sigma_{tot}$, the event is deemed to be annihilation and the particle is removed from the simulation; otherwise, it is an elastic collision and the velocity is updated by sampling $\theta$ by finding the root of $r_3 = 2\sigma_{el}^2 \int_0^\theta q \sin \theta d\theta$, where $r_3 = U[0, 1]$ (the azimuthal angle $\phi$ is chosen randomly). The momentum distribution $f(k, \tau)$ results from “binning” positron momenta at each $\tau$.

Figure 2(a) is an example of $f(k, \tau)$, calculated for Ar using 50 000 positrons initially distributed uniformly in energy up to the Ps-formation threshold. [See Supplemental Material for videos [42] of the evolution of $f(k, \tau)$ for all noble gases.] The initial distribution is seen to quickly evolve ($<100$ ns amag) to a strong Gaussian-like peak near the minimum in $B(k)$, $k_{\text{min}} \sim 0.3$ a.u., which then evolves rather slowly, producing the kneelike feature. This bunching effect becomes less effective as one moves through the sequence from He to Xe, since the minimum in $B(k)$ becomes less pronounced (with the exception of Ne, which has the deepest minimum). However, even in Xe the formation of a peak in $f(k, \tau)$ at small $\tau$ is evident. Since for any realistic initial distribution most positrons will have initial momenta $k > k_{\text{min}}$, such bunching should be expected, making the overall cooling times somewhat insensitive to the exact form of the initial distribution (see number $r_1 = U[0, 1]$ is drawn, and a collision is deemed to occur if $r_1 < P = W\Delta \tau$, where $W = v_s\sigma_{el}(E_{CM})$ is the probability rate of annihilation or elastic collision, with $\sigma_{tot} = (\sigma_{el} + \sigma_{\text{ann}})$, subject to the requirement that $P = W\Delta \tau \ll 1$ [41]. Here $\sigma_{el}$ is the elastic-scattering cross section, which is determined from the integral of the differential cross section $q = d\sigma_{el}/d\Omega = |f(\theta)|^2$, where $f(\theta)$ is the scattering amplitude for scattering angle $\theta$, which is calculated using MBT [see Eq. (31) of Ref. [24]].

MBT [24], with $\ell > 2$ partial waves described by the leading $k^2$ term in the expansion [35,36], $B(k)$ exhibits a minimum for all atoms in the sequence He to Xe [Fig. 1(a)], which becomes less pronounced as one moves through the sequence (with the exception of Ne, which has the deepest minimum). As we will see, this leads to “trapping” of positrons and slows down the cooling process in this momentum range.

The annihilation cross section in many-electron targets is parametrized as $\sigma_{el} = \pi r_0^2 Z_{\text{eff}}^2 c/v_s$ [38,39], where $r_0$ is the classical electron radius, $c$ is the speed of light, $v_s$ is the positron speed relative to the target, and $Z_{\text{eff}}$ is the dimensionless effective number of electrons that contribute to the annihilation process. Positron-atom and positron-electron correlations can result in $Z_{\text{eff}}$ being greater than the actual number of valence electrons on which positrons predominantly annihilate [20–24]. $Z_{\text{eff}}(k)$ has been calculated recently for the noble gases via MBT for $s$, $p$, and $d$-wave positrons, taking full account of correlations [24]. The $s$, $p$, and $d$ waves provide sufficient accuracy to model positron cooling below the Ps-formation threshold: although the contribution of $f$-wave positrons to the total annihilation rate is expected to be $\sim 10\%$ near the Ps-formation threshold for Xe, we will see that annihilation is unimportant until the positrons have cooled close to the minimum in $B(k)$, at which point the $f$-wave contribution is expected to be negligible ($\sim 1\%$): we note that $Z_{\text{eff}}(k) \sim k^{2\ell}$ as $k \to 0$ [24]. From He to Xe, $Z_{\text{eff}}$ becomes increasingly large and strongly peaked at low momenta [Fig. 1(b)]. This effect is due to the existence of positron-atom virtual levels [40], signified by large scattering lengths for Ar, Kr, and Xe (see Table 1 in Ref. [24]).

**Calculation of $f(k, \tau)$**—The momentum $k(\tau_i)$ of an individual positron is determined over an equidistant grid in time-density $\{\tau_i\}$ with step size $\Delta \tau$ as follows. The velocity of a gas atom is sampled from the Maxwell-Boltzmann distribution at 293 K. Both it and the positron velocities are transformed to the center-of-mass frame, in which the energy available for the collision is $E_{CM} = \mu v^2/2$, where $\mu$ is the reduced mass. A uniformly distributed random number $r_1 = U[0, 1]$ is drawn, and a collision is deemed to occur if $r_1 < P = W\Delta \tau$, where $W = v_s\sigma_{el}(E_{CM})$ is the probability rate of annihilation or elastic collision, with $\sigma_{tot} = (\sigma_{el} + \sigma_{\text{ann}})$, subject to the requirement that $P = W\Delta \tau \ll 1$ [41]. Here $\sigma_{el}$ is the elastic-scattering cross section, which is determined from the integral of the differential cross section $q = d\sigma_{el}/d\Omega = |f(\theta)|^2$, where $f(\theta)$ is the scattering amplitude for scattering angle $\theta$, which is calculated using MBT [see Eq. (31) of Ref. [24]]. If a collision occurs, a second random number $r_2 = U[0, 1]$ is drawn. If $r_2 < \sigma_{el}/\sigma_{tot}$, the event is deemed to be annihilation and the particle is removed from the simulation; otherwise, it is an elastic collision and the velocity is updated by sampling $\theta$ by finding the root of $r_3 = 2\sigma_{el}^2 \int_0^\theta q \sin \theta d\theta$, where $r_3 = U[0, 1]$ (the azimuthal angle $\phi$ is chosen randomly). The momentum distribution $f(k, \tau)$ results from “binning” positron momenta at each $\tau$.

**FIG. 1.** (a) Momentum-diffusion coefficient $B(k)/k_B T = k\sigma_{el}(k)m/M$ vs positron momentum $k$. (b) $Z_{\text{eff}}(k)$ calculated using MBT including $s$, $p$, and $d$-wave positrons annihilating on valence and inner valence subshells of the noble gases. Vertical dashed line: thermal positron momentum $k_{\text{th}} = \sqrt{3k_B T} \sim 0.0528$ a.u. at $T = 293$ K.

**FIG. 2.** (a) Density plot of positron momentum distribution $f(k, \tau)$ for Ar, normalized as $\int f(k, \tau)dk = F(\tau)$, the fraction of positrons surviving (dashed-dotted line), calculated using 50 000 positrons initially distributed uniformly in energy up to the Ps-formation threshold. Also shown is the rms momentum (black dashed line) and thermal momentum $k_{\text{th}} = \sqrt{3k_B T} \sim 0.0528$ a.u. at $T = 293$ K (solid line). (b) Fraction of positrons surviving at time-density $\tau$ (solid lines). Also marked are the values of $F$ at the shoulder time $\tau_s$ (crosses) and the complete thermalization time $\tau_{\text{th}}$ (circles).
below for further details). The small number of positrons with initial momenta below the minimum at early times cool to thermal energies relatively unimpeded, and they form a second, much smaller peak around thermal momentum $k_{th} \sim \sqrt{3k_B T} \approx 0.0528$ a.u. As the time increases, the Gaussian-like part of the distribution traverses the minimum, roughly maintaining its shape as it does so. As more positrons cool below the minimum, the two peaks in the distribution merge, eventually evolving towards the Maxwell-Boltzmann distribution. As seen in Fig. 2(a), all but a small fraction of positrons annihilate before the distribution thermalizes at $\tau \sim 400$ ns amg. This fraction is even smaller in other noble gases [see Fig. 2(b)] and has important consequences for the interpretation of measured lifetime spectra (see below). We define the “complete” thermalization time $\tau_{th}$, as the time-density at which the rms momentum of the distribution is within 1% that for a Maxwell-Boltzmann distribution at 293 K $k_{th} \sim \sqrt{3k_B T}$. The values of $\tau_{th}$ are marked in Fig. 2(b) and are presented in Supplemental Table I [42], alongside the fractions of initial positrons remaining at that time-density $F(\tau_{th})$. This fraction is a mere $F(\tau_{th}) = 0.11$ for He and reduces by more than an order of magnitude for Xe. Perhaps most remarkably, the fraction of positrons surviving to thermalization in Ne is practically zero. In Ne, cooling effectively stalls at the minimum of $B(k)$ [see Fig. 1(a)], with positrons eventually succumbing to annihilation (in spite of a relatively small $Z_{eff} \sim 6$) before they can cool further.

Comparison with experiment.—Figures 3(a)–3(e) show $Z_{eff}(\tau)$ for He, Ar, Kr, Xe, and Ne, obtained in calculations that excluded or included particle loss due to annihilation, for positrons initially distributed uniformly in energy, and with initial energy equal to the Ps-formation threshold. The monoenergetic distribution is unphysical but provides an upper limit on the cooling time. For all the noble gases, the increase in $Z_{eff}(k)$ as $k \rightarrow 0$ results in the evolution of $Z_{eff}(\tau)$ through a transient “shoulder” region resulting from epithermal annihilation at $k < k_{min}$ [1–3] towards its steady-state thermal value $\bar{Z}_{eff} \equiv \int_0^\infty Z_{eff}(k)f_T(k)dk$, where $f_T(k)$ is the Maxwell-Boltzmann distribution at $T = 293$ K. Comparisons between theory and experiment are focused around the shoulder, which is somewhat insensitive to the initial momentum distribution [5,30] [due to the bunching around the minimum in $B(k)$]. The traditional measure of the thermalization time in positron-gas studies is the “shoulder length” $\tau_s$, defined via $Z_{eff}(\tau_s) \equiv Z_{eff} - 0.1\Delta\bar{Z}$, where $\Delta\bar{Z} = Z_{eff} - Z_{min}$ and $Z_{min}$ is the minimum of $Z_{eff}(\tau)$ [43]. The calculated $\tau_s$ are given in Supplemental Table I [42], along with

FIG. 3. $\bar{Z}_{eff}(\tau)$ for He to Xe calculated using $f(k,\tau)$ excluding and including loss of particles due to annihilation, initially distributed uniformly in energy (black dashed and solid lines, respectively), and including annihilation with initial energy equal to the Ps-formation threshold (magenta dotted line). Also shown for He: experiment of Coleman et al. [4] (red circles), present calculation with $\bar{Z}_{eff}$ scaled to the measured value of $\bar{Z}_{eff} = 3.94$ (blue solid line), FP calculation of Campeanu [14] (blue dashed-dotted line), and model calculations of Boyle et al. [16] scaled to $\bar{Z}_{eff} = 3.94$ (green dash-dash-dotted line); Ar: experiments of Coleman et al. [4,8] (red circles) and FP calculation of Campeanu [8] (blue dashed-dotted line); Kr and Xe: experiment of Wright et al. [7] (red circles) and FP calculations of Campeanu [9] (blue dashed-dotted lines); Ne: experiment of Coleman et al. [4] (red circles) and FP calculation of Campeanu [8] (blue dashed-dotted lines). The calculated lifetime spectrum [i.e., observed annihilation rate $A(\tau) = -dF(\tau)/d\tau$] for Ne (blue staircase) is also shown (in arbitrary units), compared with experiment [4] (red circles). Black and red arrows mark the calculated and experimental shoulder lengths $\tau_s$. 203403-3
experimental and previous theoretical results. We now consider the results for each atom in turn, postponing the discussion of Ne as it is atypical.

**Helium.**—The calculated $Z_{\text{eff}}(\tau)$ is seen to be insensitive to both the initial distribution and whether depletion of the distribution is included or not. It is known that the MBT slightly underestimates the thermal $Z_{\text{eff}}$ in He, predicting a value of $Z_{\text{eff}} \approx 3.79$ compared with the experimental value of 3.94 [4]. Scaling the calculated $Z_{\text{eff}}(\tau)$ to the long-time steady-state experimental value, we find excellent agreement with the experiment around the shoulder region, with the calculated shoulder length $\tau_s = 1839\ ns$ amg agreeing to within 5% of the experimental value of $\tau_s = 1700 \pm 50\ ns$ amg. The overall shape and length of the calculated shoulder are in better agreement with the experiment than the FP calculation of Campeanu and Humberston [14], who used their Kohn-variational calculated cross sections. The recent diffusion-model calculation of Boyle et al. [16] is also shown. It relied on a carefully tuned model polarization potential and a zeroth-order $Z_{\text{eff}}(k)$ scaled by enhancement factors and produced $\tau_s = 1618\ ns$ amg. Overall, there is good agreement between the present calculation, that of Boyle et al., and the experiment. This complements the excellent agreement between the MBT and variational calculations and measurements of the elastic-scattering cross sections [24].

**Argon.**—The present calculated $Z_{\text{eff}}(\tau)$ are sensitive to the initial distribution at small times, but the overall cooling times for both distributions are similar. The result is weakly sensitive to whether depletion of the distribution is included or not. The calculated thermal $\bar{Z}_{\text{eff}} = 26.0$ is close to the value of 26.77 measured by Coleman et al. [4]. The calculated shoulder time $\tau_s = 369\ ns$ amg is in excellent agreement with the measured value of $362 \pm 5\ ns$ amg [4], with reasonable overall agreement in the shape of the shoulder. A much smaller shoulder length was measured in the Al Quradawi experiment, which was suspected to have suffered from the presence of impurities [10]. The present calculations show better agreement with the experiment than the FP calculation of Campeanu [8], which used the polarized-orbital cross section [20].

**Krypton.**—The calculated shoulder length and $\bar{Z}_{\text{eff}}$ are in excellent agreement with the experiment of Wright et al. [7] (fluctuations at long times are due to the small number of positrons remaining). The FP calculation of Campeanu [9], which used the polarized-orbital cross sections [21], underestimates $\bar{Z}_{\text{eff}}$.

**Xenon.**—The case of Xe is special because of the strong peaking of $Z_{\text{eff}}$ at small $k$, which means that annihilation successfully competes with cooling at epithermal positron momenta. When positron depletion due to annihilation is neglected, the positron cooling is fast ($\tau_s \sim 150\ ns$ amg), and $\bar{Z}_{\text{eff}}$ plateaus at $\sim 450$. Including depletion brings the shoulder region and shoulder length into excellent agreement with the experiment [7] (fluctuations are due to the small fraction of positrons remaining). The FP calculation of Campeanu is in serious disagreement with the experiment and the MBT result. It used the model polarized-orbital cross sections of McEachran [21], which predict elastic-scattering cross sections and $Z_{\text{eff}}(k)$ that are smaller than the MBT calculation and experiment [24].

The present calculations show that $\bar{Z}_{\text{eff}}$ is highly sensitive to the loss of particles due to annihilation. The vigorous increase in $Z_{\text{eff}}(k)$ as $k \to 0$ leads to a quasi-steady-state long-time distribution whose low-momentum component is found to be suppressed relative to the Maxwell-Boltzmann one and a steady-state annihilation rate $\dot{Z}_{\text{eff}}(\tau \to \infty) \sim 350$ that is significantly reduced from the calculated true thermal $\bar{Z}_{\text{eff}} \sim 450$. The present results thus conclusively explain the discrepancy between the gas-cell measurement of $\bar{Z}_{\text{eff}} \sim 320$ of Wright et al. [7] and the Penning-Malmberg trap measurement $\bar{Z}_{\text{eff}} \sim 401$ of the Surko group [32], whose setup ensures positrons are well thermalized.

We remark that by adding small amounts of a lighter, low-$Z_{\text{eff}}$ gas, e.g., He or H$_2$, to Xe, Wright et al. measured an increase of the pure Xe $\bar{Z}_{\text{eff}} \sim 320$ to $\bar{Z}_{\text{eff}} = 400-450$ [7], which is broadly consistent with the present calculated value and the Surko-group measurement. The mechanism for such an increase with an admixture of He is, however, unclear, given that momentum transfer is more effective in Xe as $k \to 0$ [see Fig. 1(a)].

**Neon.**—The calculated shoulder time (in this case calculated excluding the loss of particles due to annihilation) is $\tau_s = 12000\ ns$ amg, by which time the fraction of positrons remaining is practically zero [see Fig. 3(e)]. It is drastically longer than the measured value $\tau = 2700\ ns$ amg [4] (note that the FP calculation of Campeanu [8] is also slower and has not reached a steady-state value). This serious disagreement is in spite of the good agreement of the MBT elastic-scattering cross section with the experiment, including at the Ramsauer minimum [24]. Moreover, the present calculation is consistent with that expected from mass scaling the He result (which has a similar sized $\sigma_\tau$) $\tau_{\text{th}} = M_{\text{He}}/M_{\text{Ne}} \sim 5\tau_{\text{th}}^\text{He} \sim 14500\ ns$ amg.

The experimental shoulder length was determined by Coleman et al. [4] via straight line fits to the lifetime spectrum. However, in spite of the serious discrepancy in $\tau_s$, the calculated and measured lifetime spectra are in surprisingly good agreement [see Fig. 3(f)]. Importantly, the calculated $\tau_s = 12000\ ns$ amg and $\tau_{\text{th}} \sim 21000\ ns$ amg are much longer than the 0–8000 ns amg considered in the experimental analysis. As seen in Fig. 2(b), at $\tau \lesssim 8000\ ns$ amg the vast majority of positrons have already annihilated, after cooling had effectively stalled around the minimum in $B(k)$. Since $Z_{\text{eff}}(k)$ is a reasonably flat function around the minimum, a signal of many to all of the positrons annihilating at momenta close to the minimum would be observed as a leveling off of $\bar{Z}_{\text{eff}}(\tau)$, which could have erroneously been interpreted as the true thermal $\bar{Z}_{\text{eff}}$. A second possible source of error in the experimental
determination of the shoulder length is that it was affected by the presence of impurities. It is known that positron cooling in gases like CO$_2$ and N$_2$O is fast, e.g., 0.1 ns amg [10]. The presence of even minute amounts of impurities could thus lead to a significant reduction in the cooling time.

**Summary.**—Positron cooling in noble gases has been simulated using accurate cross sections calculated *ab initio* from many-body theory. The fraction of positrons surviving to thermalization was shown to be strikingly small. For Xe, the time-varying dimensionless annihilation rate $Z_{\text{eff}}(\tau)$ is shown to be strongly affected by the depletion of positrons, conclusively explaining the long-standing discrepancy between gas-cell [7] and trap-based [32] measurements in Xe. Overall, the use of the accurate atomic data gives the best agreement to date with experiment for all noble gases except Ne, the experiment for which is proffered to have suffered from incomplete knowledge of the fraction of positrons surviving to thermalization and/or the presence of impurities. New lifetime-spectra measurements, or alternatively measurements of the time-varying annihilation $\gamma$ spectra [44], are now warranted.

Data relating to this article can be accessed online [45].

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26. MBT was also applied to positron interactions with hydrogenlike ions [27] and to study the effect of positron-atom correlations on positron-molecule $\gamma$-ray spectra [28,29].
31. D. G. Green (to be published).
36. Higher-order terms in $\delta_\ell$ for $\ell > 2$ [37] contribute negligibly to $\sigma_\ell$, e.g., < 3% in Xe at the Ps-formation threshold, quickly reducing to < 1% at $k < 0.55$ a.u., and can thus be neglected.
41. In practice, we demand that $P = W \Delta \tau < 0.1$.
42. See Supplemental Material at [http://link.aps.org/supplemental/10.1103/PhysRevLett.119.203403](http://link.aps.org/supplemental/10.1103/PhysRevLett.119.203403) for (i) a table comparing the present calculated thermalization times and shoulder lengths with previous calculated and measured values for He, Ne, Ar, Kr, and Xe, and (ii) links to videos showing evolution of various quantities, e.g., the positron momentum distribution, during the cooling process for He, Ne, Ar, Kr and Xe.
45. [http://doi.org/10.17034/9da6ab49-3a41-4042-90ed-f7e479d2f437](http://doi.org/10.17034/9da6ab49-3a41-4042-90ed-f7e479d2f437)