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Orientation and alignment of the first excited p state in Li+He and Na+He scattering

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Orientation and alignment parameters for the first excited p state of Li and Na in collisions with He through direct excitation from the ground state are studied theoretically in the energy region up to $E_{\text{c.m.}} = 100$ keV by using a quasi-one-electron theory. Scattering states are expanded in terms of molecular orbitals, which are calculated by using the pseudopotential method and include electron translation factors. The approach appears to work well for Li+He, giving good agreement for the $2p$ excitation probability and orientation. For alignment, the situation is less clear because of difficulty in experimental measurement. Two-electron effects and cascades from more highly excited states cause our description of Na+He collisions to be less satisfactory. However, agreement with the experimental $3p$ excitation probability and orientation parameters where all data are available is fairly good at lower energies ($E_{\text{c.m.}} < 1$ keV) and at larger impact parameters ($b > 1.25$ a.u.).

I. INTRODUCTION

Over the last few years it has become possible to measure the orientation and alignment of the electronic charge distributions associated with atomic excited states resulting from collisions.[1] Transition probabilities and alignment and orientation parameters, studied as specific functions of impact parameter and energy, provide a complete representation of the information that characterizes the collision dynamics in detail. Studies of quasi-one-electron collision systems such as those consisting of alkali-metal atoms and helium atoms, where only the valence electron of the alkali-metal atom is considered to be active, offer us an opportunity to carry out a relatively clean test of the theories without significant complications due to electron capture channels or cascading effects arising from excitation of more highly excited states.

Two recently published experimental papers[2,3] provide alignment and orientation parameter data for lithium and sodium atoms excited by collisions with helium. At least one theoretical study[4] on alignment and orientation parameters in Na+He collisions has been attempted in order to provide a theoretical interpretation to the measurements.[2,3]

In this paper we report a study of the capability of semiclassical scattering techniques to explain some of the newest experimental results. The systems of interest are a helium atom colliding with a lithium or sodium atom. We treat as quasi-one-electron systems; the He, Li^+, and Na^+ core electrons are not considered to be active. The valence electron can occupy the ground state or excited states of the alkali-metal atoms, but we do not model charge exchange or any two-electron process. Upon excitation, we are interested in the orientation and alignment of the first excited p state, i.e., Li($2p$) or Na($3p$). By orientation we mean the imbalance in the population of the $m = \pm 1$ magnetic substates of the excited state, with the quantization axis chosen perpendicular to the scattering plane. Alignment is the angle between the incident particle velocity and the major axis of the excited-state electric charge distribution.

In the configuration interaction (CI) calculation used to determine the molecular energies and wave functions, the potential seen by the single active electron is represented by replacing each atomic core by a pseudopotential. The dynamical coupling matrix elements are calculated from the molecular orbitals (MO's) with the inclusion of atomic-orbital-type (plane-wave) electron translation factors (AO ETF's).[5] The scattering calculation is then carried out in a semiclassical impact-parameter formalism using straight-line trajectories.

Section II outlines the theory and gives details of the calculations. In Sec. III, results are presented for the excitation probabilities and the orientation and alignment parameters. Section IV contains our conclusions.

II. THEORY AND CALCULATIONS

A. Molecular states and scattering

This work is an extension of the previous quasi-one-electron H+He study[6] by this group, and closely follows the previously described theory. The closed-shell electrons of each core are assumed to be tightly bound and not to participate actively in the collision. Therefore molecular wave functions and energies were found from one-electron modified valence bond CI calculations.[5] The Schrödinger equation in the Born-Oppenheimer approximation is

$$[-\tfrac{1}{2}\nabla_r^2+V_A(\mathbf{r}_A)+V_B(\mathbf{r}_B)$$
$$+V_{\mathrm{CT}}(\mathbf{R})-E_i(R)]\phi_i(\mathbf{r},\mathbf{R})=0\ , \quad (1)$$

where $\mathbf{r}_A$ and $\mathbf{r}_B$ are the position vectors of the valence electron with respect to the He atom A and the alkali-metal core B, respectively, and $\mathbf{R}$ is the separation vector between A and B. The origin for the electronic coordinates $\mathbf{r}$ is taken to be the center of mass of the nuclei. The effective interaction potentials $V_{A(B)}$ are modeled by combining Gaussian l-dependent pseudopotentials[7,8] and polarization terms. In particular, the effective potentials of Pascale[9] are used for the He, Li^+, and Na^+ cores. Also given by Pascale[9] is V_{CT}, a three-body (cross term) interaction potential that gives the polarization of core B due to core A and the active electron.

Equation (1) is solved by using a standard CI computer code in which $\phi_i(\mathbf{r},\mathbf{R})$ is expanded in Slater-type orbitals (STO's) centered upon the alkali-metal atom B. Table I lists the STO exponents used for both systems. Figure 1 shows the calculated adiabatic energies for the states used in the scattering calculations for both Na+He and Li+He. As described in Ref. 5, the time-dependent scattering wave function, in the semiclassical treatment, is expanded on an ETF modified-molecular-state basis.[10] The ETF's are one-center, AO-type factors[5,10] centered on the alkali-metal core and having no adjustable parameters. The coupled equations represented by the interaction picture[11] are solved in a rotating coordinate frame centered on the He atom, the z axis aligned with the alkali-metal core, and the y axis perpendicular to the scattering frame. For Li+He, eight MO states have been used in the expansion of the scattering wave function ($2s\sigma$, $2p\sigma$, $3s\sigma$, $3p\sigma$, $3d\sigma$, $2p\pi$, $3p\pi$, and $3d\pi$) to include excitation up to the Li($n=3$) manifold. Fifteen MO states were retained for Na+He ($3s\sigma$, $3p\sigma$, $4s\sigma$, $3d\sigma$, $4p\sigma$, $5s\sigma$, $4d\sigma$, $4f\sigma$, $5p\sigma$, $3p\pi$, $3d\pi$, $4p\pi$, $4d\pi$, $4f\pi$, and $5p\pi$) to allow for excitation up to the Na($n=4,5$) manifolds.

B. Orientation and alignment parameters

Solutions of the time-dependent coupled equations yield the scattering amplitudes of the MO states in the rotating molecular frame. A p state has two complex amplitudes a_σ and a_π corresponding to the $np\sigma$ and $np\pi$ MO states, respectively, where n is the principal quantum number. From the state amplitudes we can calculate the parameters[12–15]

$$\lambda=\frac{|a_\sigma|^2}{|a_\sigma|^2+|a_\pi|^2}\ , \tag{2a}$$

$$\chi=\arg\left[\frac{a_\pi}{a_\sigma}\right]\ , \tag{2b}$$

where $\mathcal{P}=|a_\sigma|^2+|a_\pi|^2$ is the excitation probability of the np state. A more physical set of parameters can be defined from the Stokes parameters,[13–15] which are defined by

$$P_1=2\lambda-1\ , \tag{3a}$$

$$P_2=-2[\lambda(1-\lambda)]^{1/2}\cos\chi\ , \tag{3b}$$

$$P_3=2[\lambda(1-\lambda)]^{1/2}\sin\chi\ , \tag{3c}$$

$$P=(P_1^2+P_2^2+P_3^2)^{1/2}\ , \tag{3d}$$

where $P=1$, because in this pure $2p$ excitation model we exclude cascade effects from higher-lying states. The orientation $\langle L_\perp\rangle$ and the alignment angle γ are defined by

$$\langle L_\perp\rangle=-P_3/P \tag{4a}$$

and

$$\tan(2\gamma)=P_2/P_1\ . \tag{4b}$$

When $P=1$, all observable information about an np state can be described by $\mathcal{P}$, $\langle L_\perp\rangle$, and γ.

TABLE I. Slater-type orbital basis-set exponents used for molecular-orbital calculations of Na+He and Li+He.

Li		Na		Na	
Orbital	Exponent	Orbital	Exponent	Orbital	Exponent
$1s$	0.412	$2s$[a]	3.093	$3d$	2.375
$2s$	1.610		2.126		0.338
	0.732		0.608		0.193
	0.300		0.441	$4s$	0.282
$2p$	2.013	$2p$	0.866	$4p$[b]	0.337
	0.501		0.519	$4d$	0.235
$3s$	0.375		0.303	$4f$	0.259
$3p$	0.313	$3s$	0.441	$5s$	0.307
$3d$	0.701		0.282	$5p$	0.231
	0.333	$3p$	0.303	$6s$	0.221
			0.237		

[a] Na $2s$ through $4s$ from Pascale (Ref. 33).

[b] Na $4p$ through $6s$ from Kumar (Ref. 34).

To compare our results with those of other theoretical studies, more detailed information is needed. In particular, for comparison to the most relevant theoretical results,[4,16,17] we need the probabilities for each magnetic substate in the "natural frame,"[18,19] where the z axis is perpendicular to the scattering plane and the x axis has the same direction as the initial velocity. Given the molecular frame components of some np state, the natural frame components $|np_m\rangle$ are given by[4]

$$\begin{bmatrix} |np_0\rangle \\ |np_{+1}\rangle \\ |np_{-1}\rangle \end{bmatrix}^{\mathrm{NF}} = 2^{1/2} \begin{bmatrix} 0 & 0 & 2^{1/2} \\ -1 & -i & 0 \\ 1 & -i & 0 \end{bmatrix} \begin{bmatrix} |np\Sigma\rangle \\ |np\Pi^+\rangle \\ |np\Pi^-\rangle \end{bmatrix}^{\mathrm{MF}} , \tag{5}$$

where NF and MF stand for the natural frame and for molecular frame, respectively. The initial scattering state, with both atoms in their ground states, has positive reflection symmetry; therefore the states with negative reflection symmetry, $|np\Pi^-\rangle^{\mathrm{MF}}$ and $|np_0\rangle^{\mathrm{NF}}$, are forbidden, and their amplitudes can be set to zero.

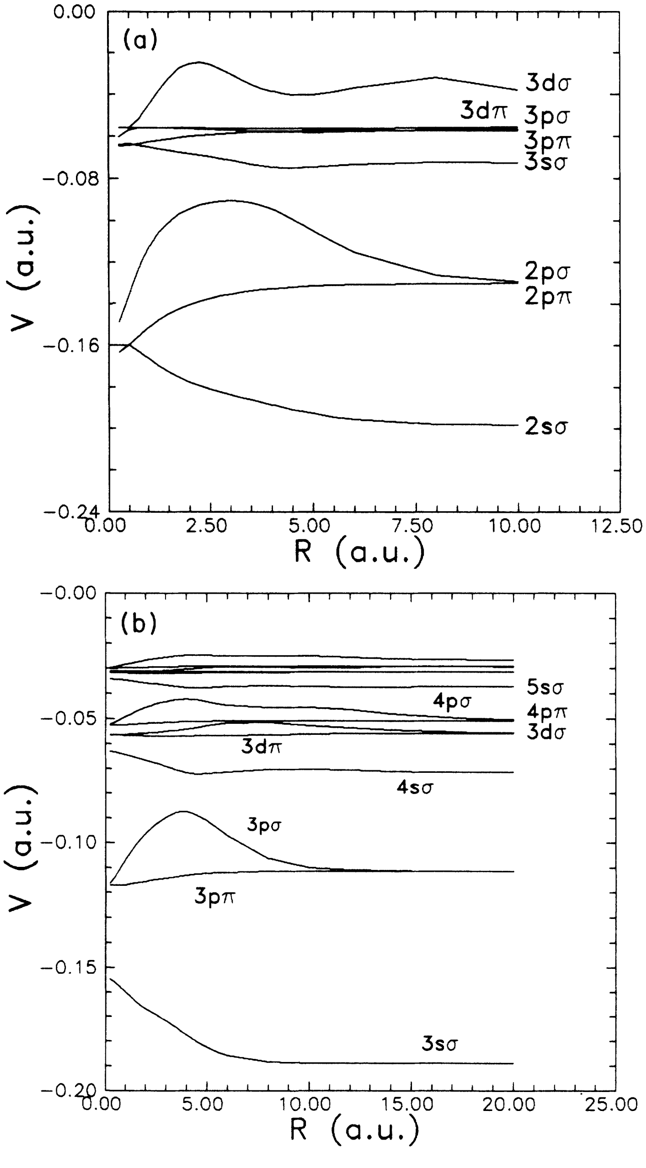

FIG. 1. Adiabatic energies used in the scattering calculations for (a) Li+He and (b) Na+He. The usual short-range core-core term, which provides a repulsive wall, is not included.

Given the NF scattering amplitudes, which obey the frame transformation of Eq. (5), the np excitation probability is $\mathcal{P}=|a_{+1}|^2+|a_{-1}|^2$, and the orientation and alignment parameters are[16]

$$\langle L_\perp \rangle = (|a_{+1}|^2 - |a_{-1}|^2)/P \ , \tag{6a}$$

$$\gamma = [\pi + \arg(a_{-1} a_{+1}^*)]/2 \ . \tag{6b}$$

The alignment angle γ is measured from the direction of the incident velocity of the alkali-metal atom.

III. RESULTS

The important collision mechanisms can be classified into two categories:[5] those that are dominant at small impact parameters ($b<1$ a.u., close collision) and those dominant at large impact parameters ($b \geq 1$ a.u., distant collision). Each mechanism is controlled by a characteristic coupling scheme and hence a characteristic excitation mechanism. For example, in a distant collision (at large impact parameters), the perturbation is weak, and a dynamical process that involves a single electron is dominant. This mechanism is properly included in the present treatment. For a close collision, on the other hand, a violent encounter causes strong mixing of a large number of states including multielectron excitation channels due to interpenetration of the atomic cores. Multielectron effects are not included in the present model. Here may be an appropriate place to compare and contrast the coupling schemes and hence the excitation mechanisms for the Li+He and Na+He systems. The important radial coupling matrix elements between $2s\sigma$ and $2p\sigma$ for Li+He and $3s\sigma$ and $3p\sigma$ for Na+He are comparable in magnitude at their peaks, at $R \cong 4.5$ a.u. However, the Li+He system shows a strong additional peak in the radial coupling around $R=0.5$ a.u. due to the stronger mixing of the $2s\sigma$ and $2p\sigma$ states at $R \leq 1$ a.u. as compared to that of the $3s\sigma$ and $3p\sigma$ states of the Na+He system in the same R region [see Figs. 1(a) and 1(b)]. This observation supports our findings of a larger $2p\sigma$ contribution to the total Li($2p$) excitation probability, amounting to more than 43% at $b \leq 1.5$ a.u. and $E \leq 10$ keV, while that for the $3p\sigma$ contribution to the total Na($3p$) excitation probability ranges from 8% at $E=0.3$ keV to 43% at $E=3.8$ keV, both at $b=1.7$ a.u. The shapes of the dominant rotational couplings, $2s\sigma$-$2p\pi$ for Li+He and $3s\sigma$-$3p\pi$ for Na+He, are rather similar while magnitudes are slightly different at $R \leq 5$ a.u. These small differences in the radial and rotational couplings are, in part, responsible for differences between the alignment and orientation parameters for the two systems.

A. Li+He

Excitation cross sections in Li+He collisions have been measured[20-23] and calculated[24] with reasonable agreement. More recently, polarization parameters were examined.[3,17,25,26] Two papers have reported detailed studies of orientation and alignment in Li+He scattering, one theoretical,[17] using a small-scale (three-state) AO method, and the other experimental.[3] Our study aims to make an extensive comparison with these two studies.

Figures 2(a)–2(c) illustrate our results for (a) Li(2*p*) excitation probabilities, and (b) and (c), orientation and alignment, respectively, of the Li(2*p*) excited state versus impact parameter for several scattering energies. The Li(2*p*) excitation probability increases with energy up to approximately 1 keV. Beyond 1 keV, the Li(2*p*) probability decreases, to the benefit of Li(2*s*) and the more highly excited states as well. In this regard, agreement with the theoretical results of Nielsen and Andersen[17] is qualitatively reasonable. The orientation below 4 keV is near zero at small *b*, and decreases to -1 (total orientation in the $m=-1$ state) in the region 2 a.u. $< b <$ 4 a.u. However, as the energy increases, the orientation at small *b* approaches -1.0 and decreases in magnitude at larger *b*. In contrast, alignment [shown in Fig. 2(c)] clearly is less sensitive to energy both in magnitude and dependence on *b*.

The calculated excitation probability, orientation, and alignment for Li(2*p*) at $b=0.93$ a.u. for varying energies are shown in Fig. 3 along with the experimental results of Andersen and Pedersen.[3] The excitation probability is displayed in Fig. 3(a). Figure 3(b) compares the calculated orientation with the experimental results.[3] The agreement is generally very good except at the lowest energies.

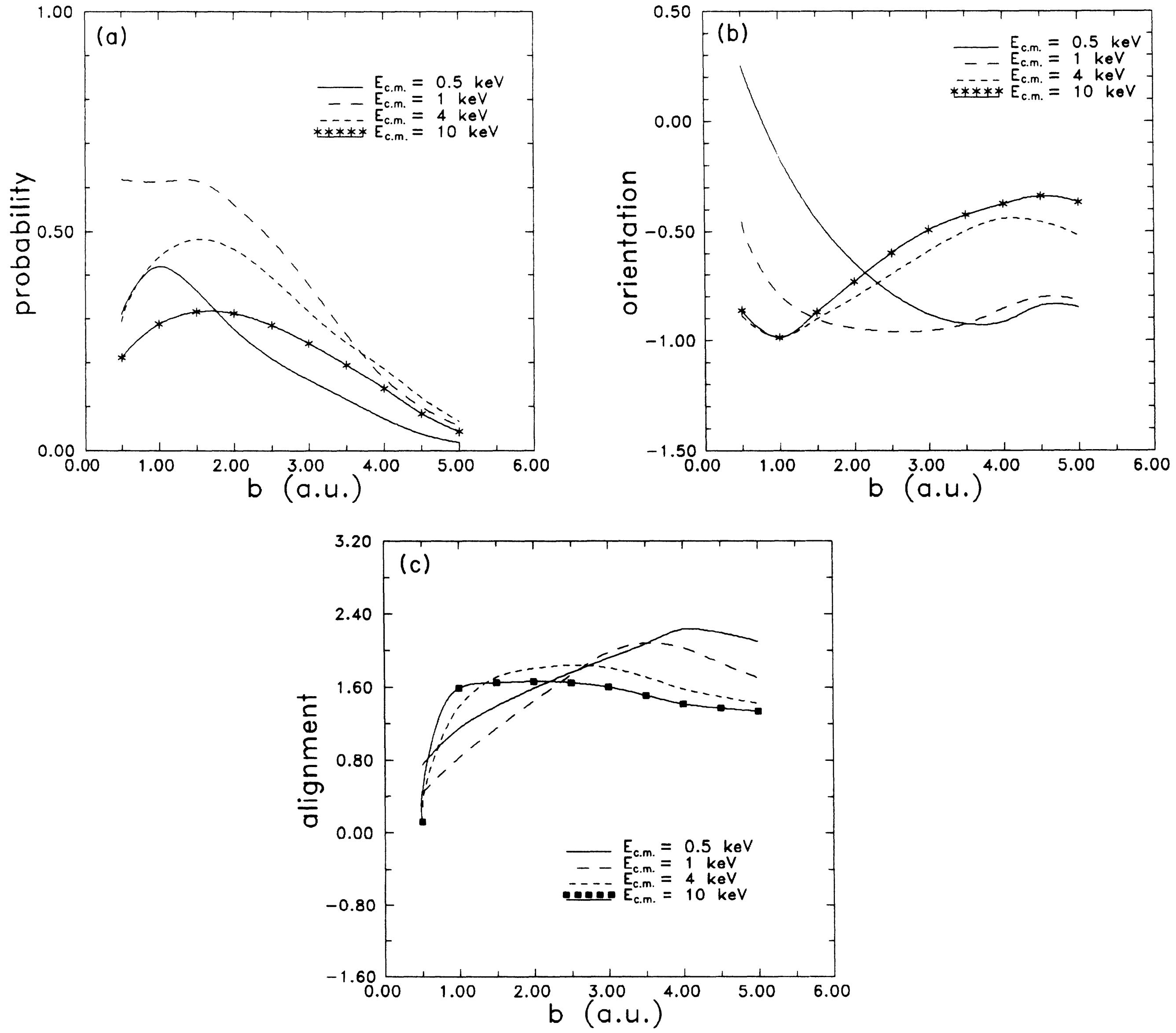

FIG. 2. Li+He scattering results for $E_{\text{c.m.}}=0.5$, 1, 4, and 10 keV and varying impact parameter. The probability for Li+He $\rightarrow$Li(2*p*)+He is shown in (a), and the orientation and alignment (in radians) of Li(2*p*) are shown in (b) and (c), respectively.

As the scattering energy is increased, the calculated orientation approaches -1.0, as is expected from the "rolling ball" model[18] based on the idea that the orientation depends on whether the trajectory of the projectile is effectively repulsive or attractive and the propensity rule,[16] but the experimental orientation increases to -0.8, presumably because of cascading from higher excited states. When an electron cascades down from a higher excited state, it usually loses orientation at each stage; thus, with a large number of cascades, the orientation approaches zero. This explains the experimental departure from -1.0. At lower energies, the calculated orientation increases, while the measured value stays nearly constant, close to -1.0. Since we expect the theory to be more reliable at low energies, this deviation is somewhat puzzling. However, at small internuclear separation below 0.5–0.6 a.u., the pseudopotential method is not expected to provide accurate potentials particularly for repulsive parts of the potentials. This problem may result in an inaccurate estimation of couplings. In addition, the straight-line trajectory that is used neglects the effect of bent trajectories. These trajectory effects become increasingly important at lower energies and may well be partly responsible for the discrepancy.

The calculated and measured alignments, shown in Fig. 3(c), exhibit nearly the same energy dependence, but the former is slightly larger. This discrepancy is somewhat disturbing. However, when the orientation is close to -1.0, the Li($2p$) electron occupies a nearly pure $m=-1$ substate, with a charge density that corresponds to a very slightly distorted toroid around the z axis. The distortion of the toroid is very small when the orientation is nearly -1, but it is this distortion that defines the major axis, thus the alignment. The alignment in such a case is very difficult to measure accurately.[27] Hence the

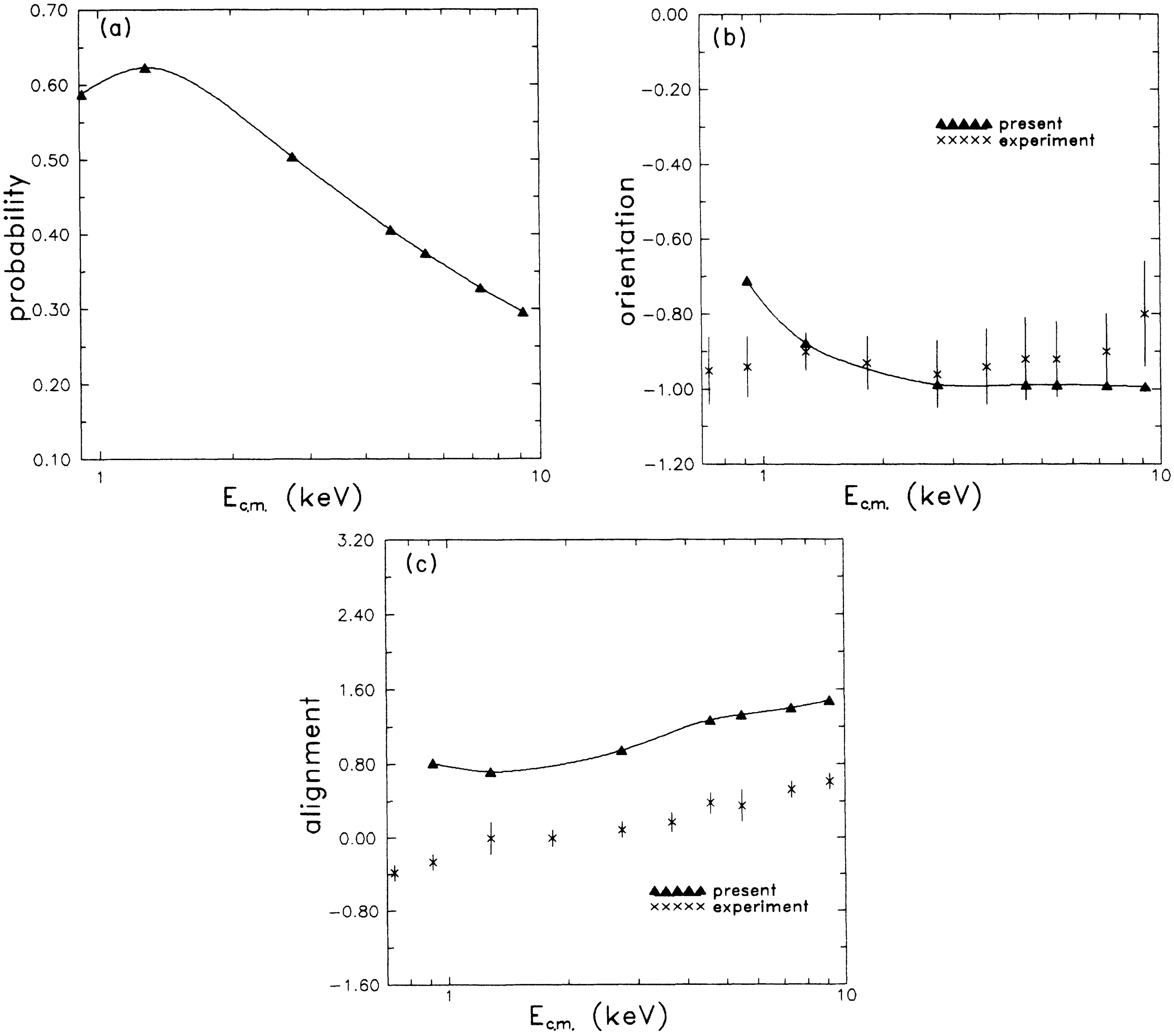

FIG. 3. Comparison of present theoretical and experimental (Ref. 3) Li+He results at $b=0.93$ a.u. The excitation probability for Li($2p$)+He is shown in (a), and the orientation and alignment (in radians) are shown in (b) and (c), respectively.

experimental result is more open to question in this case.

The comparisons of calculated and measured results for $b=1.25$ a.u. are shown in Fig. 4. Our calculated excitation probability for Li($2p$), given in Fig. 4(a), is close to the experimental result[3] for $E_{\text{c.m.}} \leq 1$ keV, but it is larger by as much as 20% as the energy increases to $E_{\text{c.m.}}=4.4$ keV. The calculated orientation, compared with the experimental result in Fig. 4(b), shows reasonable agreement except at the highest energy measured. As we observed for $b=0.93$ a.u., both the experimental orientation and the excitation probability are slightly smaller in magnitude than ours, strongly suggesting that the experimental results include cascading from higher excited states, especially at the highest energy. The calculated orientation below 1 keV is in better agreement with the measurement than is the case for $b=0.93$ a.u. [see Fig. 3(b)]. The calculated alignment, shown in Fig. 4(c), again is satisfactory in shape, but less so in magnitude. Our previous comment about the difficulty of measuring the alignment when the orientation is near -1.0 applies here as well.

B. Na+He

The excitation of Na in collisions with He has received detailed attention from both experimentalists[2,28] and theorists,[4,16,24,29] with the most recent works[2,4,16] addressing orientation and alignment in the energy range 3–13 keV. In this section, we will be particularly interested in comparing our results with the theoretical results of Andersen and Nielsen,[16] who used the three-state AO method, and Wahnon, Salas, and Courbin,[4] who included

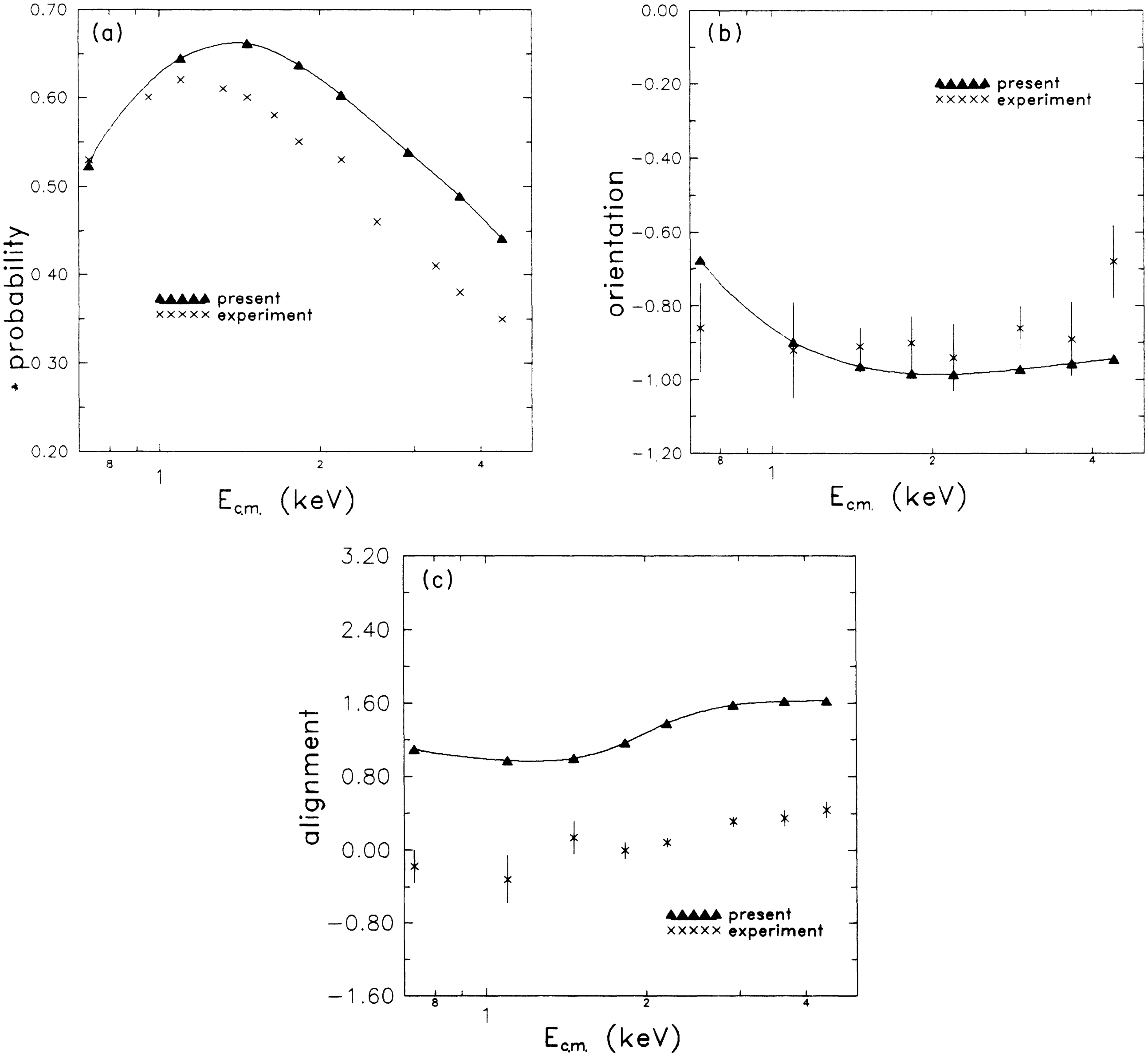

FIG. 4. Comparison of present theoretical and experimental (Ref. 3) Li+He results at $b=1.25$ a.u. The theoretical excitation probability for Li($2p$)+He is shown in (a), and the orientation and alignment (in radians) are shown in (b) and (c), respectively.

two-electron processes within a molecular representation.

Previous studies[28,29,30] have found that two-electron effects are important at intermediate energies with impact parameters below 1.3 a.u., thus invalidating the quasi-one-electron approximation at small impact parameters. However, the degree to which the approximation fails remains an open question.

In Fig. 5, we compare the present calculated Na(3*p*) probability [Fig. 5(a)], and orientation [Fig. 5(b)] and alignment parameters [Fig. 5(c)] with those measured by Andersen *et al.*[2] for $b = 1.73$ a.u. The agreement is excellent for the probability for Na(3*p*) excitation, but less so for the orientation. The most serious discrepancy occurs above $E_{\mathrm{c.m.}} = 1.5$ keV, where this calculation predicts almost total orientation, whereas the experimental orientation tends toward zero. The present results, which predict that the orientation should be approximately -1.0 for $E_{\mathrm{c.m.}} \gtrsim 2$ keV at $b \gtrsim 2$ a.u., are in accord with rolling ball model of Hertel[18,31] and with the "propensity rule" proposed by Andersen and Nielsen.[16] In Ref. 2 the experimental increase (decrease in magnitude) in the orientation is attributed to partial population of the Na(3*p*) state by cascades from higher excited states. Figure 5(a) shows the sum of the calculation probabilities for Na excitation to other excited states, i.e., states above Na(3*p*). Indeed, the summed probability of the excited states above Na(3*p*) rises to nearly 10% by 1.5 keV, suggesting an important contribution of a cascade effect. To actually calculate the cascade effect on the orientation requires detailed knowledge of the branching ratios of Rydberg

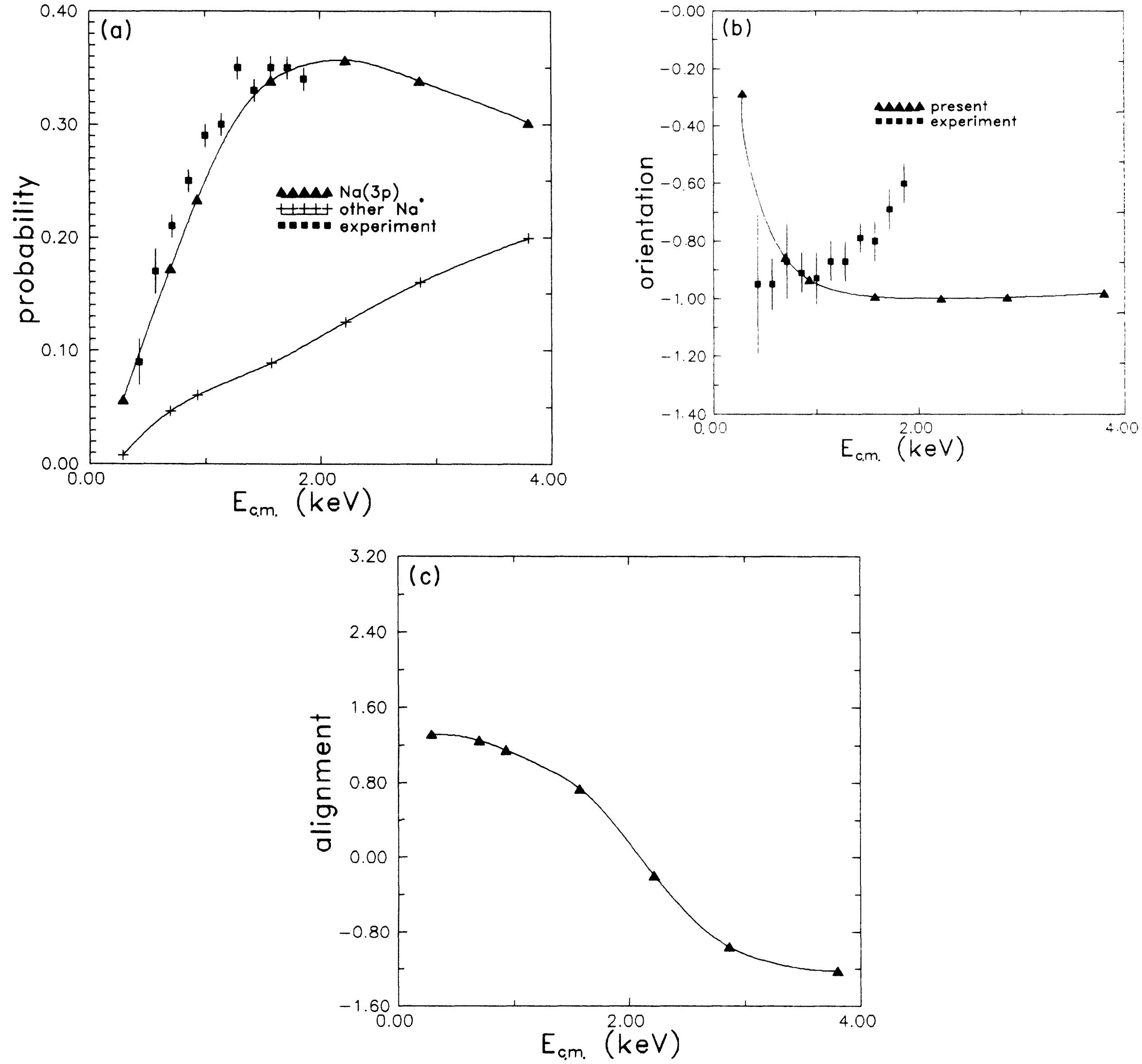

FIG. 5. Na+He scattering results for $b = 1.73$ a.u., comparing the present results with experiment (Ref. 2). The theoretical excitation probability for $\mathrm{Na+He \rightarrow Na(3}p\mathrm{)+He}$ is shown in (a), along with the sum of the probabilities for the Na 4*s*, 3*d*, 4*p*, 5*s*, 4*d*, 4*f*, and 5*d* states. The orientation and alignment (in radians) of Na(3*p*) are shown in (b) and (c), respectively.

excitations, excitation cross sections, and polarization of the excited states;[32] such a calculation will await a later study. Our calculated alignment has been included in Fig. 5(c) for completeness, although, to the best of our knowledge, no measurements now exist.

Wahnon, Salas, and Courbin[4] recently carried out an eight-state MO calculation *without* ETF but including pseudostates of He^- to allow for the excitation of He and the charge transfer of the active electron from Na to He, simulating ionization. Their calculated orientation[4] for $b=2.2$ *a.u.* is in apparent agreement with experiment[2] for $b=1.7$ *a.u.*, even *without* the inclusion of cascade effects. Because the orientation is sensitive to the impact parameter and different impact parameters were used in each case, it is difficult to assess the degree of agreement. In addition, Wahnon, Salas, and Courbin[4] did not include

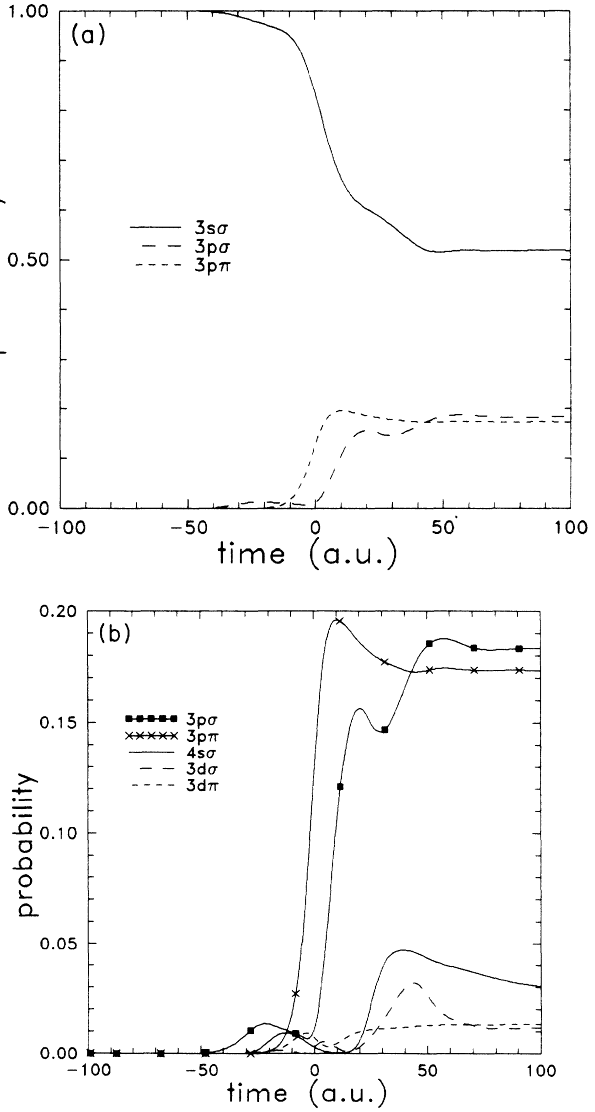

FIG. 6. Time evolution of the Na+He probabilities for $E_{\mathrm{c.m.}}=2.22$ keV and $b=1.73$ a.u. The lowest three Na states are shown in (a) and the next five states in (b).

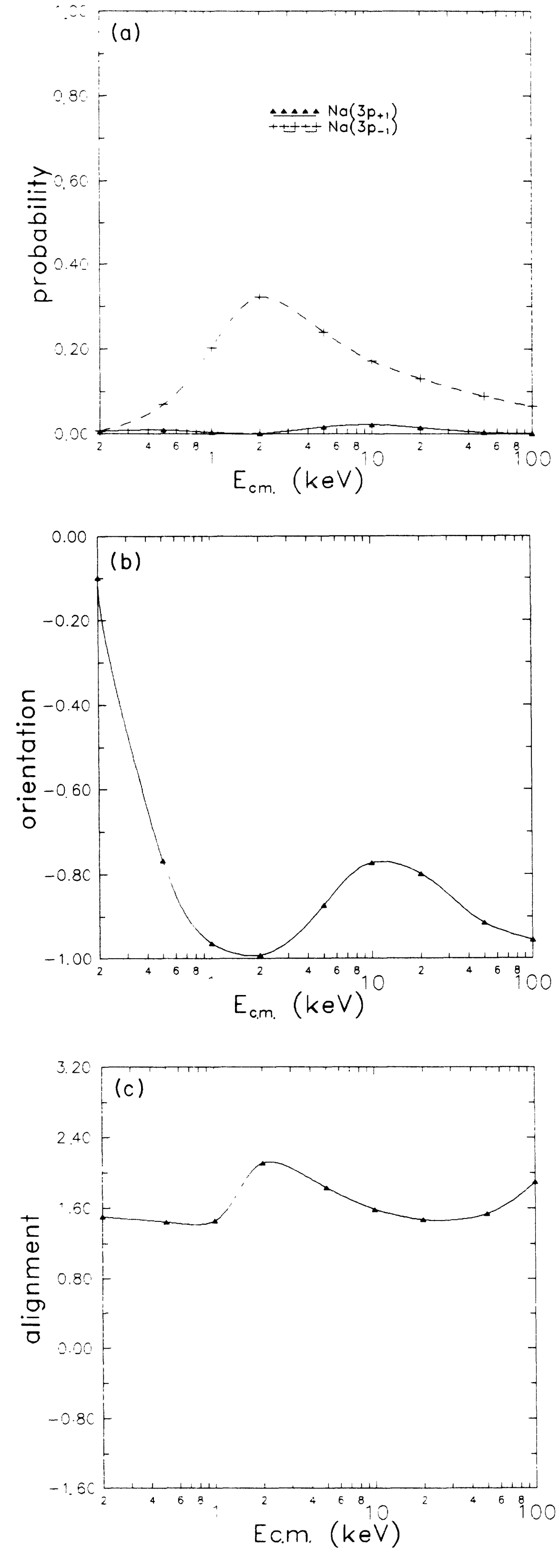

FIG. 7. Na+He scattering results for $b=2.2$ a.u. for varying energy. The excitation probability for the $Na(3p_{\pm 1})$ magnetic substates in the natural frame are shown in (a), and the orientation and alignment (in radians) of the 3p state are shown in (b) and (c), respectively.

the Na(4s) state in their basis. As the time evolution of our calculated amplitudes shows (Fig. 6), the Na(4s) state, which has a lower energy than Na(3d), is at least as important as Na(3d), in our calculations. The present work indicates that exclusion of the Na(4s) state would result in an overestimation of the probabilities of both the Na(3p) and Na(3d) states. Exclusion of the Na(4s) state does not seem to significantly alter the general shapes of the probabilities, but the individual magnitudes can vary by as much as 50%.

In Fig. 7, the variations of the present calculated Na($3p_{\pm 1}$) (a) probabilities, (b) orientation, and (c) alignment with energy for $b=2.2$ a.u. are shown. Our results agree fairly closely with the three-state AO calculations of Andersen and Nielsen (not shown).[16] However, a significant different occurs at energies above 50 keV, where our calculated $3p_{-1}$ cross section does not go to zero as theirs does, with the result that our orientation returns to -1.0 while theirs approaches zero. Wahnon, Salas, and Courbin[4] state that their results for Na($3p_{+1}$) probability and orientation significantly differ from those of the three-state calculation of Andersen and Nielsen[16] (and thus from the present results). For $b=2.2$ a.u. a specific difference between the present results and those of Wahnon, Salas, and Courbin[4] is their prediction of a large probability for Na($3p_{+1}$) excitation at all energies, resulting in the rapid increase in orientation to $+1.0$ for $E_{\text{c.m.}} > 50$ keV. The present calculated orientation de-

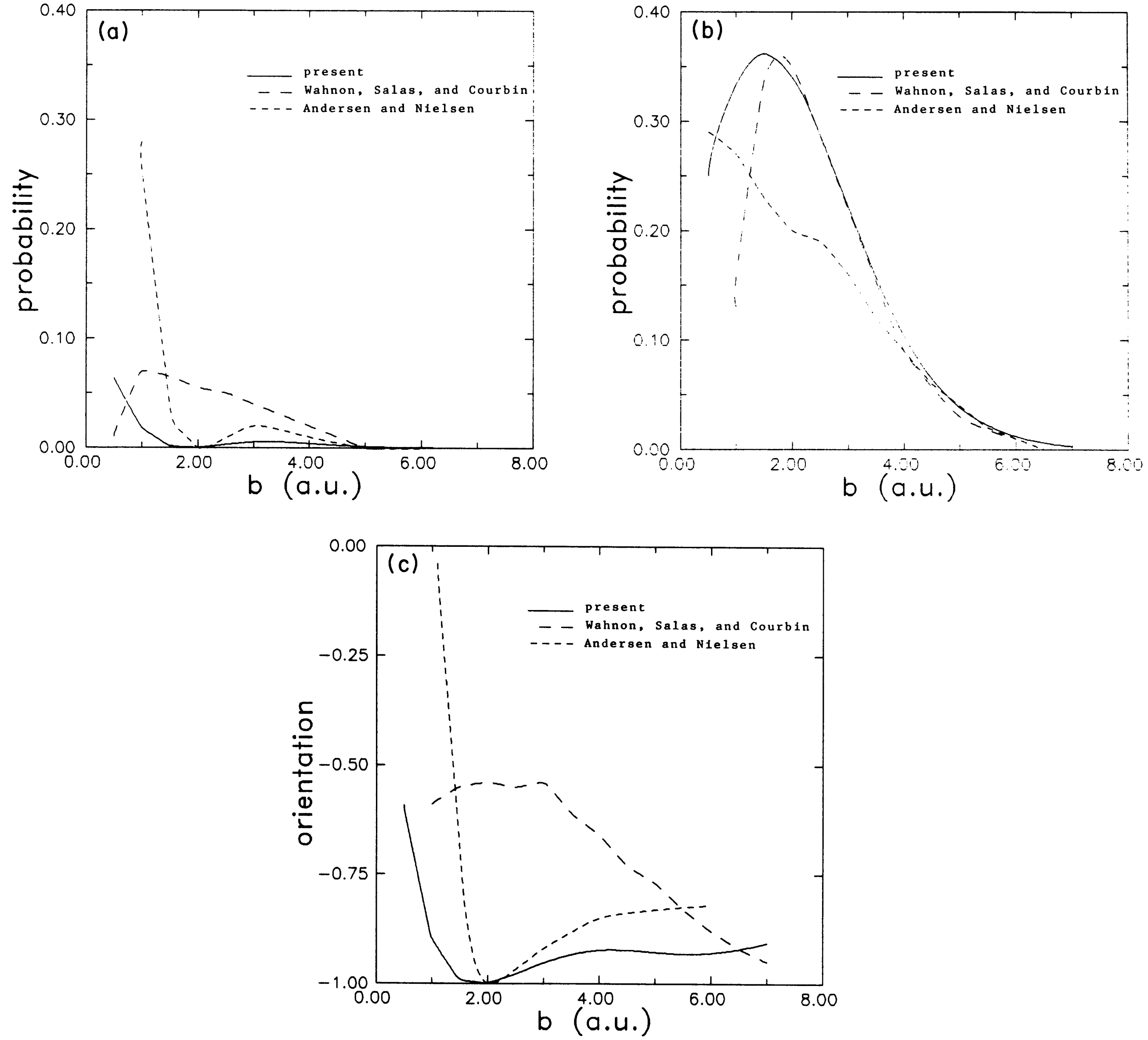

FIG. 8. Na+He scattering results for $E_{\text{c.m.}}=2.0$ keV for varying b compared to other theoretical results (Refs. 4 and 16). The excitation probability is shown for the Na($3p_{+1}$) magnetic substate in (a) and for the Na($3p_{-1}$) substate in (b). The orientation is shown in (c). Present, ——, Wahnon, Salas, and Courbin (Ref. 4), — —, Andersen and Nielsen (Ref. 16), - - -.

creases nearly to -1.0 at $E_{\mathrm{c.m.}}=1.8$ keV, where the Na($3p_{+1}$) probability is at a minimum. The orientation increases to -0.8 at 10 keV and returns to -1.0 at higher energies. The behavior of the probabilities results in structure in the alignment, as shown in Fig. 7(c).

Figure 8 shows the $3p_{+1}$ and $3p_{-1}$ probabilities [Figs. 8(a) and 8(b)] and orientation [Fig. 8(c)] for the three different theoretical treatments for varying impact parameter and $E_{\mathrm{c.m.}}=2.0$ keV. The present calculated results for all three quantities at 2.0 keV are in reasonable agreement with the three-state AO calculation of Andersen and Nielsen,[16] but not with the calculations of Wahnon, Salas, and Courbin.[4] For the $3p_{+1}$ probability, both the present result and that of Andersen and Nielsen[16] show a minimum at $b\cong 2$ a.u., while that of Wahnon, Salas, and Courbin[4] displays a maximum at $b\cong 1$ a.u. and decreases monotonically at larger b. All three calculations of the Na($3p_{-1}$) excitation probability as a function of impact parameter are in qualitative agreement, at least for $b\geq 3$ a.u., but the peak seen in the present results and those of Andersen and Nielsen[16] was not found by Wahnon, Salas, and Courbin.[4] The orientation calculated by Wahnon, Salas, and Courbin[4] is approximately -0.6 for $b<3$ a.u. and approaches -1.0 for $b>3$ a.u. This is very different from the present calculated orientation shown in Fig. 8(c), which approaches -1.0 at $b=2$ a.u. and decreases in magnitude for $b>3$ a.u. As the scattering energy increases, our calculation predicts population of the

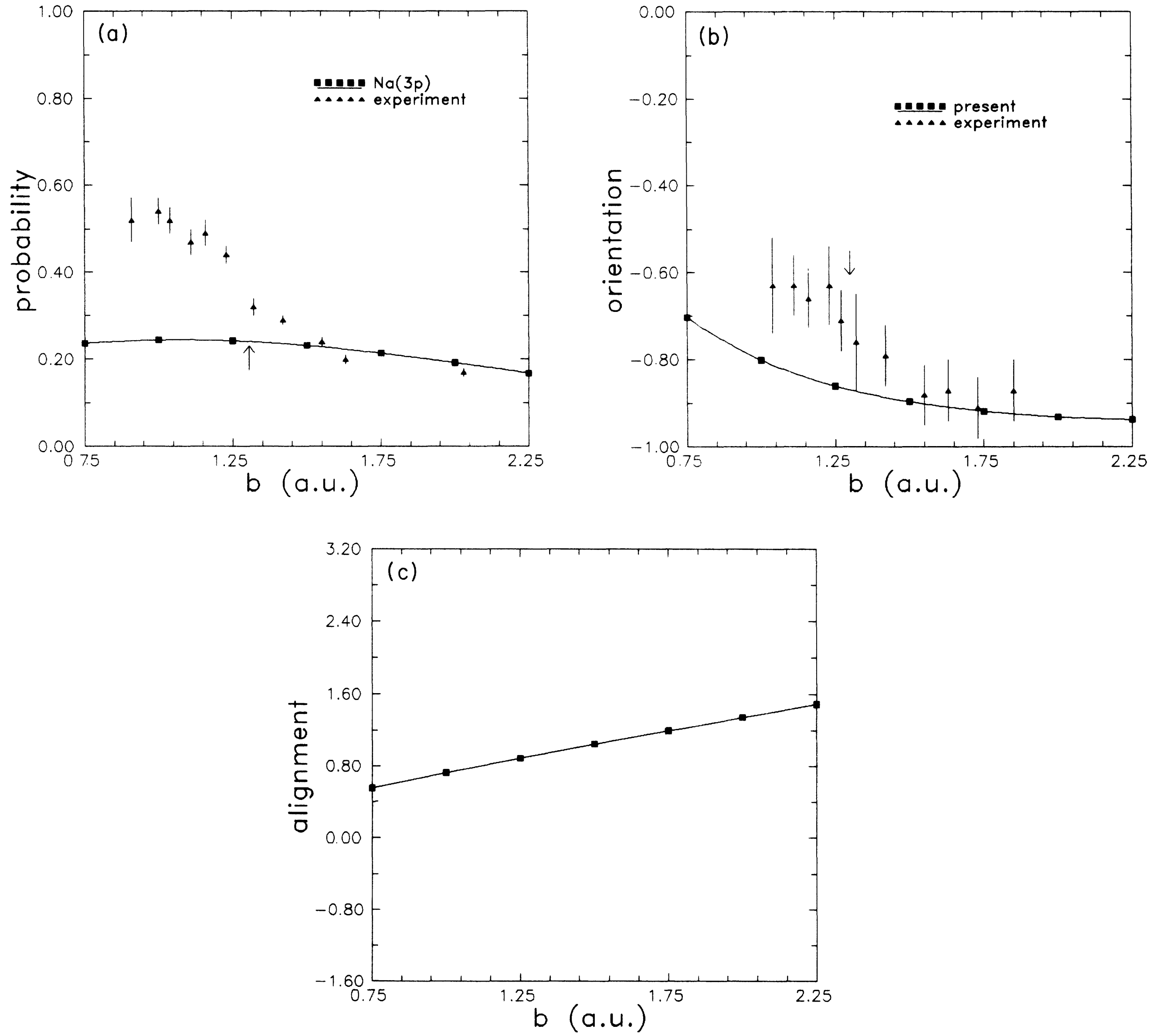

FIG. 9. Na+He scattering results for $E_{\mathrm{c.m.}}=0.86$ keV for varying b, compared to experiment (Ref. 2). The Na($3p$) excitation probability is shown in (a), and the orientation is in (b). The arrows in (a) and (b) indicate the highest impact parameters for probable He excitation. The alignment (in radians) is shown in (c).

higher Na excited states, whereas the calculation of Wahnon, Salas, and Courbin[4] predicts population of the He$^-$ pseudostates.

A final comparison with the measured probability and orientation[2] is shown in Fig. 9 for $E_{\text{c.m.}}=0.86$ keV. For $b<1.4$ a.u., the present calculated results clearly do not agree with the measurements. Below $b=1.4$ a.u., the experimental probability increases rapidly with decreasing b, while the present results increase only slightly. This divergence strongly suggests[2,28,30] that two-electron processes leading to doubly excited states and ionization play an important role in Na(3p) excitation at small impact parameters. For $b>1.5$ a.u., both the probability for Na(3p) excitation and the orientation agree very well with the experimental results,[2] indicating that only the single-electron process is involved in Na(3p) excitation. The present calculated alignment is included in Fig. 9(c) for completeness; no experimental results are available.

IV. CONCLUSIONS

The excitation of Li(2p) in Li+He collisions is a good test case for calculating orientation and alignment with a quasi-one-electron theory. This model suggests that there are no significant two-electron effects and only small, but non-negligible, cascade effects in the energy range of interest. Thus the pseudo-one-electron theory provides a satisfactory method for calculating excitation probabilities and the orientation, while the situation is less clear for alignment because sufficiently reliable measurements are lacking.

At first glance, excitation of Na(3p) in Na+He collisions also seems to be a good test case of the quasi-one-electron theory for calculating orientation and alignment parameters. However, measurements[2,30] have shown complicated behavior in the Na+He system. The quasi-one-electron model seems to fail for impact parameters below 1.5 a.u. because of the abrupt onset of double excitation[30] (Na*+He*) and ionization[2] (Na$^+$+He+e^-), as Andersen *et al.*[2] have suggested. Further studies of this system that account for double excitation and ionization appear to be necessary to explain the large Na(3p) excitation probability[2] at $E_{\text{c.m.}}=0.86$ keV and $b<1.5$ a.u. For impact parameters above 1.5 a.u., experiment and theory seem to be in reasonable agreement, but comparison becomes increasingly difficult because the orientation and, presumably, also the alignment are distorted by cascading from higher excited states.

We find the fairly good agreement of the present MO calculation with the small-scale AO calculation by Andersen and Nielsen[16] (for some parameters) to be somewhat surprising, and perhaps in some cases fortuitous, because we believe that the small size of the AO basis set used by the latter authors is inadequate for convergence. With regard to the claim by Wahnon, Salas, and Courbin[4] of good agreement with the measurements, we believe that their approach neglects important contributions and therefore that the apparent agreement could be, in part, fortuitous. Additional experimental measurements could be very helpful in resolving the issue.

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