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Polarisation of Radiation in Low-Energy $\text{He}^+ - \text{He}$ Collisions

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Synopsis

We calculated the polarisations of the emission cross sections of the $2^1\text{S} - 3^1\text{P}$ and $2^1\text{P} - 3^1\text{D}$ transitions of HeI by the two-level model for the collision energy below 20 keV, attributing the cause of alignment of the magnetic sublevels to the excitation mechanism assigned in a previous paper. Qualitative agreement between the calculated polarisations and the measured ones shows that noticeable variation of the observed polarisation with increasing impact energy is ascribed to rotation of the molecular axis following the change of scattering angle. The contribution of the secondary processes and the influence of the deviation of the symmetry axis of the atom from the internuclear axis of the separating particles, to the polarisations were discussed.

1. Introduction

We have measured the spectral intensities of HeI radiated from $\text{He}^+ - \text{He}$ collisions in the energy region from 50 eV to 20 keV and have recognised that the excited levels of different orbital angular momentum and of different spin multiplicity have different profiles of excitation functions.¹⁾ In the energy region observed, the excitation of the colliding particles occurs through transition between quasimolecular states composed in colliding period. We can, in some cases, infer the path of molecular transition from the incident channel to the low-lying outgoing channel, by considering the potential crossing in the correlation diagram between the separated atom and the united

atom.

We have shown that the measured characteristics of excitation functions originate in the type of potential curve crossing and the coupling scheme of the dominant excitation process in the interested energy region.¹⁾ This fact suggests the calculations of the two-level model may well describe the excitation process, as for the dominant process. On the basis of above inference we have showed in the previous paper²⁾ (we refer it as paper I hereafter) that the two-level coupled equations solved with theoretically predicted or reasonably assumed atomic parameters reproduce the profiles of the measured excitation functions well.

The component of the orbital angular momentum along the direction of molecular axis in quasimolecular state is conserved as the

magnetic quantum number of the separated atom in the coordinate where the quantum axis coincides with the separating direction of scattered atoms (molecular frame). Hence, alignment arises on the atoms populated by the transition of a fixed coupling scheme because the selection rules of the transition specify the quantum number A in molecular state. The radiation from the atoms is also to show characteristic behaviour of polarisation on energy dependence. In the present paper, we show that the behaviours of the polarisations derived from the excitation functions in paper I explain the energy dependences of the measured polarisations of the emission cross sections qualitatively.

2. Calculations of Polarisation

The selection rules of the transitions by rotational coupling and radial coupling are $\Delta A = \pm 1$ and $\Delta A = 0$, respectively.²⁾ The incident channel is in Σ -state. Thus, the population of the outgoing Π -state by rotational excitation results in the alignment of the magnetic sublevel $M_L' = \pm 1$ of the separated atom in molecular frame, and that of the Σ -state by radial excitation results in $M_L' = 0$. If the coupling scheme of the molecular excitation is defined, we can accordingly obtain the transition probability $P_{M_L'}$ to the magnetic sublevel M_L' by calculating the transition probability to the molecular state.

We assumed that a particle goes a classical orbit of elastic scattering; the incident particle of impact parameter b is scattered by the potential energy $V(R)$ to the angle $\theta(b)$ from the beam direction in the center-of-mass system (molecular frame). This assumption is appropriate in the collision where the impact-energy is high compared with the excitation energy. The excitation cross section σ_{M_L} of magnetic sublevel M_L in the laboratory system (atomic frame), where we take the quantum axis parallel to the incident beam direction, is given by equation (1)⁴⁾

$$\sigma_{M_L} = 2\pi \int \left[\sum_{M_L'} \left\{ D_{M_L M_L'}^L(\alpha, \beta, \gamma) \right\}^2 P_{M_L'}(b) \right] b db. \quad (1)$$

Here, $D_{M_L M_L'}^L(\alpha, \beta, \gamma)$ is the transform matrix

of the rotation group associating with the rotation of the quantum axis from the direction of the molecular axis (M_L') to the ion beam direction (M_L). In the transformation of the system of axial-symmetry, two of Euler's angles, α and γ , give no contribution to D . When the direction of molecular axis coincides with the scattering angle, we can denote it by $D_{M_L M_L'}^L(\theta(b))$. The magnetic quantum number M_L' in the molecular frame in equation (1) is zero for radial excitation and ± 1 for rotational excitation.

The polarisation degree $P = (I_{//} - I_{\perp}) / (I_{//} + I_{\perp})$ of the emission observed from the direction perpendicular to the ion-beam axis is presented with the excitation cross section σ_{M_L} of equation (1), as follows.³⁾ For singlet term transition,

$$^1S - ^1P \quad P = \frac{\sigma_0 - \sigma_1}{\sigma_0 + \sigma_1} \quad (2)$$

$$^1P - ^1D \quad P = \frac{3\sigma_0 + 3\sigma_1 - 6\sigma_2}{5\sigma_0 + 9\sigma_1 + 6\sigma_2}. \quad (3)$$

The perturbation coefficient of the triplet transition, which appears in intensity calculations of polarised light, includes time-dependent term, and the fine-structure splitting ΔE_{FS} causes quantum beats. However, the fine-structure oscillations are rapid compared with the lifetime and the time resolution of the detecting system. Thus, the oscillating terms average to zero in the present study. The polarisations of the triplet transition are given, finally, as follows;

$$^3S - ^3P \quad P = \frac{15\sigma_0 - 15\sigma_1}{41\sigma_0 + 67\sigma_1} \quad (4)$$

$$^3P - ^3D \quad P = \frac{213\sigma_0 + 213\sigma_1 - 426\sigma_2}{671\sigma_0 + 1271\sigma_1 + 1058\sigma_2}. \quad (5)$$

The measured polarisations of the triplet transition are smaller than those of the singlet transition all through the observed energy region.⁴⁾ This comes from depolarisation by the fine-structure interaction mentioned above.

3. Results

3.1 Relation between potential curve crossing and behaviour of excitation function

Details of the two-level model calculations

Fig. 1 (a) Types of potential curve crossing and coupling scheme and (b) corresponding excitation functions. $\circ$: rotational coupling, $\square$: radial coupling.

and some results were presented in paper I. Crossings of the potential energy curves of the low-lying excited levels with the curve of the incident channel have been classified into two types, as shown in figure 1(a). In type I crossing, rotational coupling is a dominant transition moment and, in type II crossing, radial coupling is important. The structures of the excitation function arising from the former type interaction is mainly determined by relative position of the potential energy curve to the incident channel potential, and the excitation function (σ_{rot}) generally has a profile similar to curve I in figure 1(b) (the high-energy structure). In the latter type interaction, the transition moment of radial coupling further exerts strong influence on the profile of the excitation function in the low-energy region. When the matrix element of the transition moment localizes near the crossing point, which is the case in $\text{He}^+ - \text{He}$ collisions,⁶⁾ the excitation function (σ_{rad}) is similar to curve II in figure 1(b) (the low-energy structure). The excited level of the separated atom correlates to more than two molecular states of outgoing channels. Therefore, we may regard a real excitation function as a compound of those produced by a few excitation processes dominant in the high and low-energy regions respectively.

3.2 The measured excitation functions and polarisations

The measured excitation functions and polarisations have been presented elsewhere.^{1,4)} Those of the 3^1P and 3^1D levels are shown in

figures 2 and 3, respectively. On the cross section of the 3^1P level in figure 2(a), the effect of the rotational coupling excitation from the incoming channel $1s\sigma_g 2p\sigma_u \rightarrow 2^2\Sigma_g \rightarrow 1s\sigma_g 2p\sigma_u 3p\pi_u \rightarrow 2^2\Pi_{g1}$ is remarkable in high-energy side (high-energy structure) and rather small contribution of the $2^2\Sigma_g \rightarrow 1s\sigma_g 2d\sigma_g \rightarrow 2^2\Sigma_{g1}$ transition by radial coupling is seen in low-energy side (low-energy structure). On the other hand, the $2^2\Sigma_g \rightarrow 1s\sigma_g$

Fig. 2 (a) The measured-emission cross section of the He I 5016 Å ($2^1\text{S} - 3^1\text{P}$) line and (b) its polarisation ($(\sigma_{//} - \sigma_{\perp}) / (\sigma_{//} + \sigma_{\perp})$).

Fig. 3 (a) The measured emission cross section of the He I 6678 Å (2^1P-3^1D) line and (b) its polarisation.

$5g\sigma_g \rightarrow 2\Sigma_{g2}$ transition by radial coupling makes prominent low-energy structure, and the high-energy structure which arises from the $2\Sigma_{g0} - 1s\sigma_g 2p\sigma_u 4f\pi_u \rightarrow 2\Pi_{g2}$ transition is recognised in high-energy limit of the observed energy region on the cross section of 3^1D in figure 3(a).

3.3 The calculated excitation functions and polarisations

We calculated the excitation functions of high and low-energy structures separately by the two-level model and added them, as previously mentioned. The intensity ratio of the added functions was determined to such a value that the resulting excitation function may reproduce the profile of the measured cross section. The cross section obtained in this method is not always correct. However, the result is useful so far as we are interested in the polarisation of radiation, because the polarisation is derived from the ratio of cross sections of aligned levels (see equations 2 to 5), not directly concerning with absolute values of them.

In the calculations, we used curve V_0 in figure 2 of paper 1 as the incoming channel potential $2\Sigma_{g0}$. The observed behaviour of the

low-energy structure of the 3^1P level cross section resembles to that of the 3^1D level cross section. Thus, we adopted curve V_3 in figure 2 of paper 1 for the potential curves of both molecular states $1s\sigma_g 2d\sigma_g \rightarrow 2\Sigma_{g1}$ and $1s\sigma_g 25g\sigma_g \rightarrow 2\Sigma_{g2}$, and regarded curve f_b in figure 8 of paper 1 as coupling elements for both transitions of $2\Sigma_{g0} - 1s\sigma_g 2d\sigma_g \rightarrow 2\Sigma_{g1}$ and $2\Sigma_{g0} - 1s\sigma_g 25g\sigma_g \rightarrow 2\Sigma_{g2}$. Curve V_3 and curve f_b have been obtained by the experimental and the theoretical studies, respectively (see paper 1).

The atomic parameters necessary to calculate the high-energy structure of $n=3$ level cross section are poorly known. The potential energy of the $1s\sigma_g 2p\sigma_u 3p\pi_u \rightarrow 2\Pi_{g1}$ state dissociating to $He^+ + He(3^1,3P)$ is close to V_3 in figure 2 of paper 1 at a large internuclear distance (R), because V_3 connects to the separated atom state of $He^+ + He(3^1,3D)$. At small R , however, it probably lies between V_2 and $V_{2'}$ in figure 2 of paper 1 to produce an excitation function which resembles to the high-energy structure of the measured 3^1P level cross section (cf. figure 7 in paper 1). Contrary to the 3^1P level cross section, the high-energy structure of the measured 3^1D level cross section does not attain a maximum in the energy region observed. Hence, the potential curve of the $1s\sigma_g 2p\sigma_u 4f\pi_u \rightarrow 2\Pi_{g2}$ state dissociating to $He^+ + He(3^1,3D)$ may locate higher than the curve of the $2\Pi_{g1}$ state,

Fig. 4 Potential energy differences between $2\Sigma_{g0}$ and $2\Pi_{g1}$ (curve I) and between $2\Sigma_{g0}$ and $2\Pi_{g2}$ (curve II) assumed for the calculations of the rotational coupling excitations of the 3^1P and 3^1D levels, respectively (see text).

Fig. 5 (a) The calculated excitation functions of the 3^1P level and (b) the polarisations. σ_{rot} : the excitation cross section by rotational coupling, σ_{rad} : 1/11 of the cross section by radial coupling. $\sigma_{\text{total}} = \sigma_{\text{rot}} + \sigma_{\text{rad}}$.

Fig. 6 (a) The calculated excitation functions of the 3^1D level and (b) the polarisations. σ_{rot} : 8/10 of the cross section by rotational coupling.

far from $^2\Sigma_{g0}$, at small R . The potential energy differences between the $^2\Sigma_{g0}$ and $^2\Pi_{g1}$ states and the $^2\Sigma_{g0}$ and $^2\Pi_{g2}$ states, assumed for our calculations, are shown in figure 4. There exists no available information about the matrix elements of $\langle \Sigma_{g0} | (i/\hbar) L_{\perp} | \Pi_{g1} \rangle$ and $\langle \Sigma_{g0} | (i/\hbar) L_{\perp} | \Pi_{g2} \rangle$. We used the same function as f_b in figure 3 of paper 1, except that it decreases linearly from the value at $R=1$ to zero at $R=0$ owing to orthogonality of wave functions between Σ_{g0} and Π_{g1} or Π_{g2} , for the rotational coupling elements.

The calculated excitation functions of the 3^1P level and the polarisations of radiations from the $2^1\text{S} - 3^1\text{P}$ transition, and those of the 3^1D level and from the $2^1\text{P} - 3^1\text{D}$ transition are shown in figures 5 and 6, respectively. In figure 5(a), the total cross section (σ_{total}) was made of the cross section of the rotational coupling excitation (σ_{rot}) added 1/11 of the radial coupling cross section (σ_{rad}), and, in figure 6(a), 8/10 of σ_{rot} was added to σ_{rad} .

4. Discussion

For collisions of high-energy region, the molecular axis of the separating particles comes close to the ion beam direction because of small scattering angle. Thus, an increase in the ratio of σ_{rot} to σ_{total} generates alignment of magnetic sublevel $M_L = \pm 1$ (σ_1) in the laboratory system (atomic frame); the polarisation of the $2^1\text{S} - 3^1\text{P}$ transition converges to -1 (equation 2 and σ_{rot} in figure 5(b)) and that of the $2^1\text{P} - 3^1\text{D}$ transition converges to $3/9$ (equation 3 and σ_{rot} in figure 6(b)) at complete alignment to $M_L = 1$ level. These results explain qualitatively decreasing tendency, with increase in collision energy, of the measured polarisation of the $2^1\text{S} - 3^1\text{P}$ transition in figure 2(b) and increasing to 0.3 or more of the $2^1\text{P} - 3^1\text{D}$ transition in figure 3(b), in the high-energy region. The large scale structures appearing in the low-energy side of figures 2(b) and 3(b) probably correspond to the valley-like structures of the polarisation of σ_{rad} in figures 5(b) and 6(b), respectively.

The qualitative agreements between the behaviours of the measured polarisations and those of the calculated ones suggest that our assignment about dominant excitation processes

in each energy region is appropriate as a whole. On the other hand, there are considerable discrepancies between them; the value of the polarisation in the high-energy region of the 2^1S-3^1P transition, the collision energy where the minimum polarisation appears, the value of the minimum, and the gradient of increase in the low-energy side of the 2^1P-3^1D transition. The calculations not cited in this paper have shown that the behaviour of the polarisation is scarcely influenced, while the profile and absolute value of the excitation function considerably change, by the variation of the parameters included in the coupled equations. Thus, we consider other mechanisms which give significant influence on the behaviour of the polarisation in the following section.

4.1 Secondary processes

Secondary processes among the outgoing channels which have been neglected cause the change of molecular state populations and accordingly exert influence on the polarisations. The transitions from the excited levels which dissociate to other separated atom states different from the convergence limit of the investigating level at large R scarcely affect the population of the level, because the level is strongly coupled with the incident channel and the population is mainly determined by the transition from this channel.

Another possible secondary processes are the transitions between the levels dissociating to the same separated atom limit. In figure 1(a) the potential energy curves of V_n and V_x converge to the same separated atom limit at large R , and rotational coupling between them becomes effective.⁶⁾ The influence of this coupling does not make insecure the basis of our inference about the coupling scheme of dominant excitation process at small R , because it hardly changes the profile of an excitation function.

We have insufficient knowledge about the strength of the coupling force concerning in the populations of magnetic sublevels of $n=3$ in He^+-He collisions. Slow decrease, compared with the calculated result, of the measured polarisation of the 2^1S-3^1P transition at high-energy region in figure 2(b) may be partly due to the $\Sigma \rightarrow \Pi$ transition by this rotational coupling. On the 2^1P-3^1D transition, $\Sigma \rightarrow \Pi$ and

$\Pi \rightarrow \Delta$ transitions at high-energy region make the polarisation smaller and $\Pi \rightarrow \Sigma$ transition makes it larger and, hence, qualitative discussion gives no definite conclusion about the behaviour of the polarisation. The calculation by Grosser⁶⁾ showed that the rotational coupling at large R causes population change of 10% at most in the collisions similar to the present experiment.

In the low-energy region the mixing of Π -state accompanied with the $\Sigma \rightarrow \Pi$ transition by rotational coupling above mentioned makes the minimum value of the polarisation of the 2^1P-3^1D transition in figure 5(b) large, and the difference between the measured value of -0.4 and the calculated value of -0.2 becomes larger. Therefore, we have to consider another mechanism that produces alteration of the polarisation so as to improve the discrepancies.

4.2 Deviation of the molecular axis from the scattering direction

The direction of molecular axis is much concerned in calculation of the polarisations. In the present calculations, the direction of elastic scattering was taken as the direction of molecular axis. We also tried to calculate the excitation functions and the polarisations using the deflection function of inelastic scattering, but no significant change was recognised on the behaviours of the polarisations.

The deviation of the symmetry axis of atoms from the internuclear axis of the separating atoms causes noticeable change of the polarisation. This deviation possibly occurs by stopping of rotation of the molecular axis at the limit of the molecular interaction region; the

Fig. 7 Relation between the interaction radius and the direction of the molecular axis. b : impact parameter. θ : scattering angle, $\Delta\theta$: deviation of molecular axis from scattering angle.

body-fixed molecular state changes to the space-fixed atomic state at the interaction radius R_I and the symmetry axis of the separated atom is fixed to the direction of the molecular axis at R_I as shown in figure 7. Thus, the direction of symmetry axis of the atom depends on the size of the interaction radius. The size of R_I is, roughly speaking, as large as the size of the region where the difference between the potential energy of the molecular state and the energy of the separated atoms at large R is finite (see figure 2 in paper 1).

Figures 8 and 9 show the polarisations calculated for the high-energy structure of the $2^1\text{S} - 3^1\text{P}$ transition and for the low-energy structure of the $2^1\text{P} - 3^1\text{D}$ transition at various

Fig. 8 The calculated polarisations for various R_I , of the $2^1\text{S} - 3^1\text{P}$ transition excited by rotational coupling.

Fig. 9 The calculated polarisations for various R_I , of the $2^1\text{P} - 3^1\text{D}$ transition excited by radial coupling.

interaction radii, respectively. The measured polarisations of the low-energy structures agree much better with the calculated ones for the interaction radius of $1 \sim 2$ au. This value of the radius is nearly equal to the limit of the impact parameter where the transition probability $P(b)$ has finite value (ref. 7, figures 5 and 10 in paper 1). This fact suggests that, on the radial excitation of the 3^1D level in low-energy collisions, the direction of the symmetry axis of the separated atom closely relates to the direction of the molecular axis at the molecular excitation.

The consideration about the deviation of the symmetry axis owing to the finite size of the molecular interaction region improves also the behaviour of the calculated polarisations at the high-energy region and, however, the discrepancy between the measured polarisation of the $2^1\text{S} - 3^1\text{P}$ transition and the calculated one is still noticeable. The radiation trapping effect for the resonance line of the $1^1\text{S} - 3^1\text{P}$ transition probably affect the measured polarisation of the $2^1\text{S} - 3^1\text{P}$ transition. For high-energy collisions, the secondary process of rotational coupling at large R discussed in the preceding section becomes effective and it makes the calculated polarisation large.

5. Conclusion

It has been shown that the polarisation of the emission from the low-lying excited level has the characteristic energy dependence corresponding with the characteristic profile of the excitation function. Attributing the cause of the alignment of the magnetic sublevel of $M_L = \pm 1$ to the rotational coupling excitation and that of $M_L = 0$ to the radial coupling excitation, we calculated the polarisations of the S-P and P-D transitions by the two-level model. Qualitative agreements between the calculated polarisations and the measured ones show that there are dominant excitation processes of the specified coupling schemes and that the noticeable variation of the observed polarisation with increasing impact energy is ascribed to the rotation of the molecular axis following the change of scattering angle.

We discussed two additional processes to explain the discrepancies between the meas-

ured polarisations and the calculated results; one is rotational coupling, accompanied with the potential degeneracies at large R , among the molecular states which dissociate into the same separated atom limit and another is stopping of the molecular axis rotation at the limit of the molecular interaction region. The former process becomes effective in the high-energy region and may explain considerably large polarisation of the high-energy structure of the 2^1S-3^1P transition. The latter process gives the polarisation of large negative value in the low-energy structure by the radial excitation. The measured polarisation of the 2^1P-3^1D transition agrees on the whole with the calculated polarisation for interaction radius of about 2 au. The interatomic distance of 2 au

corresponds roughly to the radius of the interaction region of the molecular transition.

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