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Spectroscopic study of promethiumlike bismuth with an electron-beam ion trap: Search for alkali-metal-like resonance lines

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We experimentally settle the three-decades-old question of whether or not promethiumlike heavy ions have strong resonance lines in hot plasmas. In 1980, Curtis and Ellis [Phys. Rev. Lett. **45**, 2099 (1980)] predicted that promethiumlike heavy ions should have an alkali-metal structure with a ground-state configuration of $4f^{14}5s$, and should exhibit strong $5s$-$5p$ resonance lines in hot plasmas. However, after many experimental efforts, no clear indication of the predicted resonance lines was found. The work presented herein resolves this question by clearly showing that the predicted resonance lines for bismuth are negligibly weak because of the presence of the $[4f^{13}5s^2]_{7/2}$ metastable state, even though the ground-state configuration is $4f^{14}5s$, as predicted. To obtain these results, we used an electron-beam ion trap that made it possible to exploit a fine-tuned charge-state distribution, and analyzed the experimental spectra with collisional-radiative model calculations.

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The *ns-np* resonance lines in alkali-metal-like ions with a closed $(n-1)$ shell are important for fusion plasmas; their prominence can cause serious radiation loss while also being useful for spectroscopic diagnostics. Thus, the hydrogenlike, lithiumlike ($n=2$), sodiumlike ($n=3$), and copperlike ($n=4$) sequences have been extensively studied both experimentally and theoretically. Their *ns-np* resonance lines have been identified and their wavelengths have been compiled for a wide range of atomic numbers (Z) [1–5]. For $n=5$ (i.e., the promethiumlike sequence), Curtis and Ellis [6] predicted in 1980 that the alkali-metal-like $4f^{14}5s$ configuration can be achieved for elements with $Z>73$ and that experimentalists would observe the prominent $5s$-$5p$ resonance doublet lines for promethiumlike heavy ions. Following their prediction, many experimental and theoretical efforts were devoted to identifying the predicted resonance lines.

The first attempt to find the resonance lines was by beam-foil spectroscopy with gold ($Z=79$) [7,8] in the 1980's. Träbert and Heckmann [8] identified the $5s$-$5p$ doublet lines in their beam-foil spectra of gold, but the spectra contained complex features because many lines exist near the identified lines, making the identification tentative. Subsequently, a beam-foil measurement with lead ($Z=82$) was performed at RIKEN and the wavelength of the $5s$-$5p_{3/2}$ line was reported by Hutton *et al.* [9,10]. However, this identification should also be considered as tentative because the corresponding data are essentially unpublished. In 1994, Fournier *et al.* [11] identified the brightest line in their spectra of uranium ($Z=92$) obtained with a high-temperature, low-density tokamak plasma as the $5s$-$5p_{3/2}$ resonance line, while their collisional-radiative (CR) model calculations showed that lines due to the transitions to the metastable $4f^{13}5s^2$ state could be brighter than the resonance lines. This inconsistency and the complex spectra containing many lines from a broad charge-state distribution made the identification tentative.

In the 2000's, another attempt to find the resonance lines, this time using an electron-beam ion trap (EBIT) in Berlin [12], was made for tungsten ($Z=74$) [9]. An EBIT can produce and trap highly charged ions that interact with a monoenergetic electron beam, making it possible to obtain simple spectra based on narrow charge-state distributions. Thus, an EBIT is a powerful tool to observe and identify previously unreported lines [13]. In fact, the tungsten spectra obtained with the Berlin EBIT revealed simple spectra containing a few prominent lines [9,10]. However, in spite of intensive ongoing theoretical efforts [14–17], the observed spectra are still not well understood. As a result, the identification made from the Berlin EBIT spectra was also tentative. Thus, in the three decades since the prediction by Curtis and Ellis, there has yet to be a definite identification of the resonance lines.

In this Rapid Communication, we report the clear identification of promethiumlike bismuth ($Z=83$) observed by using the compact EBIT at The University of Electro-Communications in Tokyo. We also show that the $5s$-$5p$ resonance lines are negligibly weak in plasma over a wide range of electron densities, even though the ground-state configuration is $4f^{14}5s$, as predicted.

For the present study, a compact EBIT called CoBIT [18] was used to produce highly charged bismuth ions. The device consists essentially of an electron gun, a drift tube, an electron collector, and a high-critical-temperature superconducting magnet surrounding the drift tube. The electron beam emitted from the electron gun is compressed by the axial magnetic field (typically ∼0.08 T) produced by the magnet. The compressed high-density electron beam ionizes the bismuth introduced as a vapor from an effusion cell [19]. The bismuth ions are axially and radially confined by the well potential applied to the drift tube and the space charge of the high-density electron beam, respectively. The trapped ions are further ionized by successive electron-impact ionization.

Spectra from the trapped bismuth ions were observed with a grazing-incidence flat-field spectrometer [20] through an observation slit in the drift tube. The spectrometer consists of a laminar-type diffraction grating with 1200 grooves per mm (Hitachi 001-0660) and a Peltier-cooled back-illuminated CCD (Roper PIXIS-XO: 400B). For the experiments reported

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herein, no entrance slit was used because the EBIT constitutes a line source that serves the same function as that of a slit. The spectral resolution of this arrangement was typically 0.03 nm, which was mainly limited by the electron-beam width. The wavelength was calibrated by using well-known transitions in Fe XIV and Fe XV [21] measured separately from the bismuth spectra by injecting an $Fe(C_5H_5)_2$ vapor into CoBIT. The uncertainty in the wavelength calibration was estimated to be 0.01 nm.

We analyzed the spectra by using CR-model calculations with a minimal set of excited levels, consisting of 980 fine-structure levels of $4f^{14}nl$, $4f^{13}5snl$, $4f^{13}5p^2$, $4f^{13}5p5d$, $4f^{12}5s^25l$, and $4d^94f^{14}5s^2$ for promethiumlike Bi^{22+} and 251 fine-structure levels of $4f^{14}5snl$, $4f^{14}5p5l$, and $4f^{13}5s^2nl$ for samariumlike Bi^{21+} ($n = 5$ and 6, $l \leqslant 4$). With the CR model, excited-level populations were obtained by solving quasi-stationary-state rate equations that account for electron collisions (excitation, deexcitation, and ionization) and radiative decays (electric-dipole, electric-quadruple, and electric-octupole, and magnetic-dipole and magnetic-quadruple). Recombination from higher-charge states was omitted because the electron-beam energy was below the first-ionization energy of each ion. Atomic data (i.e., energy levels, electron collision strengths, and radiative decay rates) were calculated by using the HULLAC code (version 7) [22]. The HULLAC calculations are based on fully relativistic wave functions, and the collision strengths are calculated in the distorted-wave approximation.

The spectra for electron energies of 500, 580, and 640 eV are shown in Fig. 1(a). Each spectrum was acquired over a 30-min exposure. Because of the limited bandwidth of the spectrometer, the regions of shorter (12–16 nm) and longer (23–27 nm) wavelengths were measured separately. Additionally, no sensitivity correction was applied, which means that the intensities at different wavelengths cannot be compared. The lines indicated by the arrows in the spectrum at 580 eV are not observed in the spectrum at 500 eV; thus, these lines can be assigned to transitions in samariumlike Bi^{21+} because the ionization energy of europiumlike Bi^{20+} (532 eV [23] or 548 eV [24]) is greater than 500 eV. Similarly, the lines indicated by the arrows in the spectrum at 640 eV can be assigned to transitions in promethiumlike Bi^{22+} because they are not observed in the spectrum at 580 eV, which is below the ionization energy of samariumlike Bi^{21+} (608 eV [23] or 621 eV [24]). Our previous studies have shown that this assignment method is reliable [18,25,26]. Note also that 640 eV is below the ionization energy of promethiumlike Bi^{22+} (641 eV [23] or 659 eV [24]).

Spectra for promethiumlike Bi^{22+} and samariumlike Bi^{21+} calculated with the CR model are shown in Figs. 1(b) and 1(c), respectively. An electron density of 10^{10} cm^{-3}, which is typical for CoBIT [27], was assumed for the model calculation. The results in Fig. 1 indicate that the experimental results agree overall with those calculated by the CR model, although there is a slight shift in wavelength between the two. According to the present calculation, the ground state of promethiumlike Bi^{22+} is $4f^{14}5s$, as predicted by Curtis and Ellis [6]. However, the prominent lines in the CR-model spectrum of promethiumlike Bi^{22+} are not the $5s$-$5p$ resonance lines [indicated by arrows in Fig. 1(b)], but the $4f^{13}5s^2$-$4f^{13}5s5p$ transitions. Fournier *et al.* [11] performed CR-model calculations for uranium and obtained the result that the intensity of the transitions to the metastable state could be comparable with or rather brighter than that of the resonance line. On the other hand, our present calculation for bismuth shows that the intensity of the resonance lines is negligibly weak (almost vanish) compared with that of the transitions to the metastable state. This seemingly peculiar phenomenon is explained below.

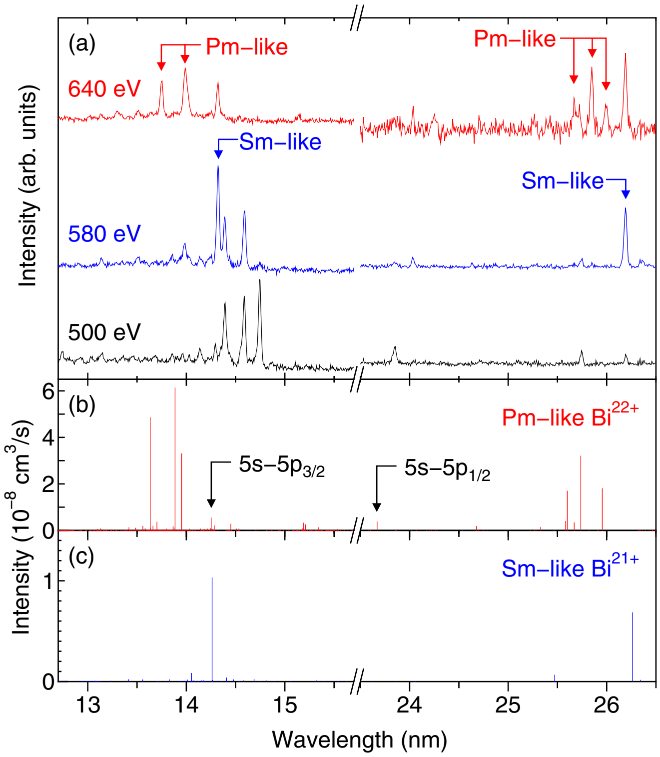

FIG. 1. (Color online) (a) Experimental spectra of highly charged bismuth ions observed at electron energies of 500, 580, and 640 eV. (b) Spectrum of promethiumlike bismuth calculated with a CR model for an electron energy of 640 eV. (c) Spectrum of samariumlike bismuth calculated with a CR model for an electron energy of 580 eV. The vertical axes in (b) and (c) correspond to the photon emissivity coefficient, which is defined as nA/n_e, where n is the fractional population of the upper level, A is the Einstein A coefficient, and n_e is the electron density. An electron density of 10^{10} cm^{-3} is assumed for both (b) and (c).

Figure 2 shows the energy levels of promethiumlike Bi^{22+}. The excitation rate from the ground-state $4f^{14}5s$ level is largest to the $4f^{14}5p$ levels, which means that the $5s$-$5p$ resonance lines should be prominent if the population of the ground state is dominant. However, because the metastable $[4f^{13}5s^2]_{7/2}$ state has a long lifetime (estimated by the present calculation to be ∼40 s), its population can dominate that of the ground state mainly through the following indirect excitation:

$$
\begin{aligned}
e_i^- + 4f^{14}5s &\rightarrow 4f^{13}5s5d + e_s^- \\
&\quad \hookrightarrow 4f^{13}5s5p + h\nu \\
&\quad \hookrightarrow 4f^{13}5s^2 + h\nu'.
\end{aligned}
$$

Actually, in the present calculation, the equilibrium population of the metastable $[4f^{13}5s^2]_{7/2}$ state is estimated to be an order of magnitude larger than that of the ground state. As a result,

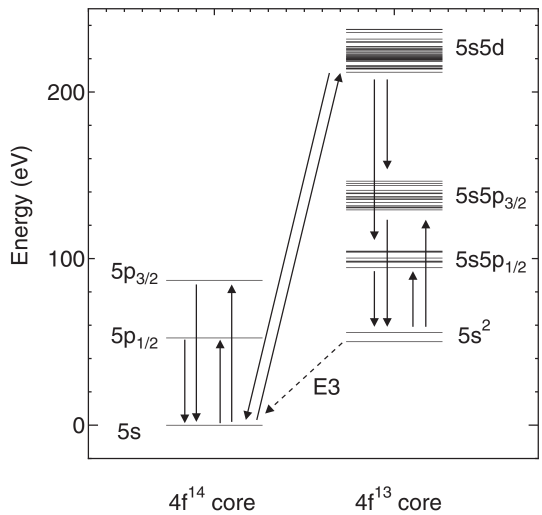

FIG. 2. Calculated energy levels of promethiumlike Bi^{22+}.

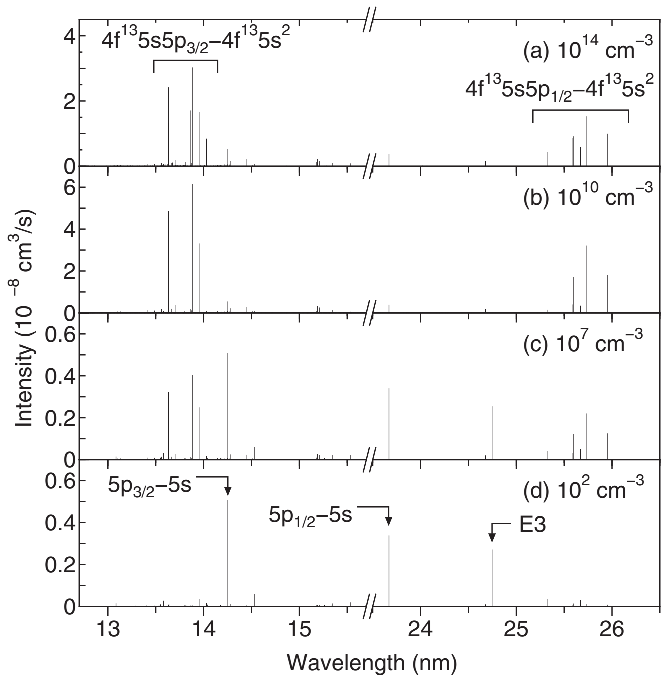

FIG. 3. Same as Fig. 1(b) but for electron densities of (a) 10^{14}, (b) 10^{10}, (c) 10^{7}, and (d) 10^{2} cm^{-3}.

the $[4f^{13}5s^2]_{7/2}$-$4f^{13}5s5p$ transitions dominate the $5s$-$5p$ resonance lines in the spectrum of the promethiumlike ion.

However, in the spectrum of samariumlike Bi^{21+}, the prominent doublet resonance lines $4f^{14}5s^2$-$4f^{14}5s5p$ are found at 14.3 and 26.2 nm. For samariumlike Bi^{21+}, all the $4f^{13}5s^25p$ excited states can decay to the ground state via electric-dipole transitions to the $4f^{14}5s5p$ levels with transition probabilities on the order of 10^4–10^8 s^{-1}. Therefore, there is no metastable state in samariumlike Bi^{21+}; as a result, the resonance lines become prominent, as observed in the present experiment, and as reproduced by the CR-model calculation. Table I lists the wavelengths of the lines in promethiumlike Bi^{22+} and samariumlike Bi^{21+} that are identified in this work. The table also lists the previous theoretical values for samariumlike Bi^{21+} by the multiconfigurational Dirac-Fock (MCDF) method [28], which show good consistency with the present result.

Because the EBIT technique provides simple spectra with a narrow charge-state distribution compared with beam-foil and plasma spectroscopies, these identifications for promethiumlike and samariumlike heavy ions are definite rather than tentative. Hutton *et al.* [9] also observed promethiumlike tungsten using an EBIT and a spectrometer with spectral resolution comparable to that of our present spectrometer, but they gave no clear identification. The complexity for tungsten is likely due to the fact that the ground state of promethiumlike tungsten is not $4f^{14}5s$ but $4f^{13}5s^2$; furthermore, the metastable $4f^{12}5s^25p$ states are near the ground state [14,16]. Note that, although Curtis and Ellis [6] predicted through nonrelativistic calculations that the transition of the ground-state configuration from $4f^{13}5s^2$ to $4f^{14}5s$ occurs between $Z = 73$ and 74, their relativistic calculation [29] and several subsequent theoretical studies [14,30,31] indicate that the transition should occur near $Z = 78$.

TABLE I. Experimental and theoretical wavelengths and theoretical transition probabilities (A) for transitions in promethiumlike Bi^{22+} and samariumlike Bi^{21+}. The notation $a(b)$ for the transition probability denotes $a \times 10^b$.

		Wavelength (nm)			A (s^{-1})	
Ion	Transition	HULLAC[a]	MCDF[b]	Expt.[a]	HULLAC[a]	MCDF[b]
Bi^{21+}	$[5s^2]_0$-$[5s5p_{3/2}]_1$	14.26	14.28	14.34	1.9(11)	1.9(11)
	$[5s^2]_0$-$[5s5p_{1/2}]_1$	26.26	26.87	26.20	6.9(9)	5.5(9)
Bi^{22+}	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{3/2}]_{7/2}$	13.63		13.77	2.1(11)	
	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{3/2}]_{9/2}$	13.89		14.0[c]	2.0(11)	
	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{3/2}]_{5/2}$	13.95			1.8(11)	
	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{1/2}]_{5/2}$	25.60		25.7[c]	7.3(9)	
	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{1/2}]_{9/2}$	25.74		25.86	7.6(9)	
	$[4f_{7/2}^{-1}5s^2]_{7/2}$-$[4f_{7/2}^{-1}5s5p_{1/2}]_{7/2}$	25.96		26.01	5.9(9)	

[a]Present results.
[b]Reference [28].
[c]Blend.

Figure 3 shows how the spectra calculated with the CR model depend on electron density. At an extremely low densities such as 10^2 cm^{-3}, where the coronal limit can be applied, the $5s$-$5p$ doublet resonance lines appear clearly, as shown in Fig. 3(d). In addition to the resonance lines, the electric-octupole (E3) transition $[4f^{14}5s]_{1/2} - [4f^{13}5s^2]_{7/2}$ appears at 24.7 nm because the collision frequency is low enough to allow the $[4f^{13}5s^2]_{7/2}$ metastable state to decay to the ground state. As the electron density increases, the intensity of the transitions from $4f^{13}5s5p$ to the metastable $4f^{13}5s^2$ state increases and becomes comparable to that of the $5s$-$5p$ resonance lines at 10^7 cm^{-3} [see Fig. 3(c)]. At 10^{10} cm^{-3}, which is a typical electron density for CoBIT, the transitions to $4f^{13}5s^2$ already dominate the $5s$-$5p$ resonance lines, as seen in Fig. 3(b). Additionally, at higher densities such as 10^{14} cm^{-3}, which are typical of fusion devices, the transitions to $4f^{13}5s^2$ are dominant, as seen in Fig. 3(a). Consequently, in the spectra of promethiumlike heavy ions, the transitions to the metastable $4f^{13}5s^2$ state should generally dominate those to the $5s$ -$5p$ resonance lines over a wide range of electron densities. Therefore, in practical fusion plasmas, the $5s$-$5p$ resonance lines would vanish. Note that the metastability (lifetime of the metastable $4f^{13}5s^2$ state) should depend on Z. In general, the metastability should become weaker (the lifetime should become shorter) for higher Z elements. Thus the present study would not deny the conclusion made by Fournier *et al.* [11], who identified the strongest line in their beam-foil spectrum of uranium as the resonance line.

In summary, by using a compact electron-beam ion trap, we observe $5s$-$5p$ transitions in promethiumlike and samariumlike bismuth. Comparing the experimental results with calculations based on a collisional-radiative model allows us to definitely identify several lines for promethiumlike heavy ions. We show that, because of the large population of the $4f^{13}5s^2$ metastable state, the alkali-metal-like $5s$-$5p$ resonance lines in promethiumlike ions are negligible in plasmas over a wide range of electron densities, even though the ground-state configuration is alkali-metal-like $4f^{14}5s$.

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[1] E. Hinnov, Phys. Rev. A **14**, 1533 (1976).
[2] J. Reader and N. Acquista, Phys. Rev. Lett. **39**, 184 (1977).
[3] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **37**, 2764 (1988).
[4] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **38**, 2699 (1988).
[5] W. R. Johnson, S. A. Blundell, and J. Sapirstein, Phys. Rev. A **42**, 1087 (1990).
[6] L. J. Curtis and D. G. Ellis, Phys. Rev. Lett. **45**, 2099 (1980).
[7] B. M. Johnson, K. W. Jones, T. H. Kruse, L. J. Curtis, and D. G. Ellis, Nucl. Instrum. Methods **202**, 53 (1982).
[8] E. Träbert and P. H. Heckmann, Z. Phys. D **1**, 381 (1986).
[9] R. Hutton, Y. Zou, J. R. Almandos, C. Biederman, R. Radtke, A. Greier, and R. Neu, Nucl. Instrum. Methods B **205**, 114 (2003).
[10] S. Wu and R. Hutton, Can. J. Phys. **86**, 125 (2008).
[11] K. B. Fournier, M. Finkenthal, S. Lippmann, C. P. Holmes, H. W. Moos, W. H. Goldstein, and A. L. Osterheld, Phys. Rev. A **50**, 3727 (1994).
[12] C. Biedermann, A. Förster, G. Fussmann, and R. Radtke, Phys. Scr., T **73**, 360 (1997).
[13] A. Komatsu, J. Sakoda, M. Minoshima, H. A. Sakaue, X.-B. Ding, D. KATO, I. Murakami, F. Koike, and N. Nakamura, Plasma Fusion Res. **7**, 1201158 (2012).
[14] M. J. Vilkas, Y. Ishikawa, and E. Träbert, Phys. Rev. A **77**, 042510 (2008).
[15] E. Träbert, M. J. Vilkas, and Y. Ishikawa, J. Phys.: Conf. Ser. **163**, 012017 (2009).
[16] V. Jonauskas, Šarūnas Masys, A. Kynienė, and G. Gaigalas, J. Quant. Spectrosc. Radiat. Transfer **127**, 64 (2013).
[17] U. I. Safronova, A. S. Safronova, and P. Beiersdorfer, Phys. Rev. A **88**, 032512 (2013).
[18] N. Nakamura, H. Kikuchi, H. A. Sakaue, and T. Watanabe, Rev. Sci. Instrum. **79**, 063104 (2008).
[19] C. Yamada, K. Nagata, N. Nakamura, S. Ohtani, S. Takahashi, T. Tobiyama, M. Tona, M. Sakurai, A. P. Kavanagh, and F. J. Currell, Rev. Sci. Instrum. **77**, 066110 (2006).
[20] H. Ohashi, J. Yatsurugi, H. A. Sakaue, and N. Nakamura, Rev. Sci. Instrum. **82**, 083103 (2011).
[21] A. Kramida, Y. Ralchenko, J. Reader, and N. A. Team, NIST Atomic Spectra Database (version 5.0), http://physics.nist.gov/asd.
[22] A. Bar-Shalom, M. Klapisch, and J. Oreg, J. Quant. Spectrosc. Radiat. Trans. **71**, 169 (2001).
[23] Present calculation.
[24] T. A. Carlson, C. W. Nestor, Jr., N. Wasserman, and J. D. McDowell, At. Data Nucl. Data Tables **2**, 63 (1970).
[25] A. Komatsu, J. Sakoda, N. Nakamura, H. A. Sakaue, X.-B. Ding, D. Kato, I. Murakami, and F. Koike, Phys. Scr., T **144**, 014012 (2011).
[26] J. Yatsurugi, E. Watanabe, H. Ohashi, H. A. Sakaue, and N. Nakamura, Phys. Scr., T **144**, 014031 (2011).
[27] N. Nakamura, E. Watanabe, H. A. Sakaue, D. Kato, I. Murakami, N. Yamamoto, H. Hara, and T. Watanabe, Astrophys. J. **739**, 17 (2011).
[28] L. J. Curtis, J. Opt. Soc. Am. B **3**, 1102 (1986).
[29] D. G. Ellis and L. J. Curtis, Nucl. Instrum. Methods **202**, 339 (1982).
[30] C. E. Theodosiou and V. Raftopoulos, Phys. Rev. A **28**, 1186 (1983).
[31] V. Kaufman, E. Träbert, P. H. Heckmann, G. Möller, V. Lodwig, and J. H. Blanke, Phys. Scr. **42**, 705 (1990).