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Author

Shirley, D.A.

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D. A. Shirley

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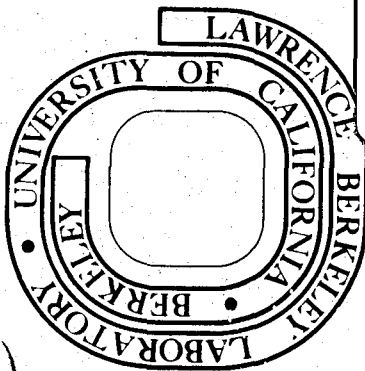
Department of Chemistry and
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

ABSTRACT

A theoretical model is described for calculating energy shifts of Auger satellites--Auger transitions that occur in the presence of a "spectator" vacancies. The shifts arise through a combination of direct and indirect (or relaxation) interactions between spectator and regular hole states. Both can be expressed in terms of two-electron Coulomb and exchange integrals, and approximate pairwise additivity of the shifts due to holes or spectator vacancies is demonstrated. An approximate method, based on the "equivalent-cores" approximation, is employed to estimate numerical values of the indirect term using only tabulated two-electron integrals for atomic ground states. Satellite shifts for the $L_3M_{45}M_{45}(M_{45})$ transition are thereby estimated for elements with $70 \leqslant Z \leqslant 100$. The predicted value of -38 eV for bismuth is in excellent agreement with the experimental value -36 ± 5 eV reported for ^{210}Bi by Haynes, Velinsky, and Velinsky. For americium the predicted value is -61 eV; Freedman and Porter reported an experimental value -56 eV. Other predicted satellite shifts in bismuth also show reasonably good agreement with less accurate experimental values. These results thus strongly support previous interpretations of satellite shifts and appear to provide a fairly accurate and very easy method for predicting these shifts.

I. INTRODUCTION

A normal Auger transition takes an atomic system from a one-hole initial state to a two-hole final state, with the energy difference going into the kinetic energy of the ejected electron. Thus the Auger energy of the transition

$$M(i)^+ \rightarrow M(jk)^{2+} + e^- \quad , \tag{1}$$

which carries element M from an initial state with a hole in the i orbital to a final state with holes in the j and k orbitals, is given by

$$E(ijk) = E(i) - E(jk) \quad . \tag{2}$$

Here $E(i)$ and $E(jk)$ are the total energies of the i and jk hole states.

Recently, Auger spectra in several heavy elements have shown Auger satellites.[1,2] These are Auger transitions that occur in the presence of additional, "spectator" vacancies which are present both before and after the transitions. An ℓ satellite on the above (ijk) transition would involve the process

$$M(i\ell)^{2+} \rightarrow M(jk\ell)^{3+} + e^- \quad , \tag{1a}$$

going from a two-hole initial state to a three-hole final state. This ℓ satellite transition would be denoted $(ijk(\ell))$, and its energy would be given by

$$E(ijk(\ell)) = E(jk\ell) - E(i\ell) \quad . \tag{2a}$$

The subject of this paper is the development of a simple theoretical scheme for estimating the shift in energy of the satellite line relative to the main

line in an Auger transition,

$$\Delta E(ijk(\ell)) \equiv E(ijk(\ell)) - E(ijk) \quad . \tag{3}$$

The scheme that will actually be used is approximate, but it is based on a rigorous analysis of the factors that lead to Auger shifts. In principle it might be possible to obtain $\Delta E(ijk(\ell))$ from self-consistent field (SCF) calculations on the appropriate one-, two-, and three-hole states (cf. Eqs. (2), (2a), and (3)). It is, however, by no means clear that this "brute force" approach is feasible at this time. Such a calculation would entail taking small differences between very large numbers. It could also have convergence problems, because an SCF calculation on a highly-excited multiple core-hole state would not be protected by the Variable Principle. Indeed there does not yet exist a comprehensive set of atomic core-level binding energies based on SCF calculations, let alone a set of Auger energies (these would involve one- and two-hole states). The present work was in fact motivated by earlier success in estimating these two quantities by approximate methods involving only calculated ground-state properties.[3-5]

The model for calculating $\Delta E(ijk(\ell))$ is developed in Sec. II. Numerical results are presented and compared with available experimental values in Sec. III. A summary is given in Sec. IV.

II. THE MODEL

The Auger satellite energy shift $\Delta E(ijk(\ell))$ can be expressed in terms of the total energies of four states by combining Eqs. (2), (2a), and (3). Barring convergence problems that might arise, it should be possible to make rather accurate SCF hole-state calculations of these total energies. As discussed above, however, the prediction of satellite shifts from calculated total energies is not a very efficient approach. It would be desirable, instead, to obtain these shifts from calculated energy <u>differences</u>. In the discussion below, which is carried out in the nomenclature of Hartree-Fock theory, it will be shown that the Auger shifts can be expressed in terms of a relatively small number of two-electron Coulomb and exchange integrals. This derivation is done in the spirit of a polarization-potential theory by Hedin and Johansson,[6] who derived a method for calculating the relaxation energy associated with ionization in terms of two-electron integrals. This approach would have no computational advantage if applied literally, because hole-state SCF calculations would still be necessary. Its advantage lies rather in the physical insight it provides by isolating the two-electron integrals that are important in hole-hole interactions. These integrals are then subdivided into two classes: those that are small enough to be safely neglected and those that must be retained. Fortunately all of the latter class can be rather well estimated by approximate methods that do not require hole-state SCF calculations.

Let us consider Fig. 1. The hole states which have a spectator

vacanvy ℓ are built up from the corresponding "regular" Auger hole states

i and jk in a three-step Gedanken experiment. The total energy difference

$E(i\ell) - E(i)$, for example, is the sum of the energy differences of the three

steps considered separately. The first step takes the system to a hypothetical

state in which an electron has been removed from orbital ℓ without taking into

account the presence of the i hole. The energy change of this process is

just the binding energy of an ℓ orbital in a free atom, $E(\ell)$.

In the second step the direct interaction between the i and ℓ

holes is added. The energy of the two-hole state is thus increased by this

repulsive interaction an amount $\mathscr{J}(i\ell)$. From standard multiplet-coupling

theory $\mathscr{J}(i\ell)$ can be expressed in terms of two-electron Coulomb and exchange

integrals. Since the resolution usually available in Auger satellite spectra

is not adequate to distinguish details of angular-momentum coupling between

hole states, it will suffice for our purposes to use average multiplet energies.

Thus, if the i and ℓ orbitals were in an s and a p shell, for example,

we could write $\mathscr{J}(i\ell)$ in terms of Slater integrals as[7]

$$\mathscr{J}(i\ell) \;=\; F^0(i\ell) - \frac{1}{6}\,G^1(i\ell) \quad , \tag{4}$$

and similarly for other cases.

The third step in Fig. 1 involves the "indirect" interaction between

orbitals i and ℓ. These are brought about through the separate

influence of the holes in orbitals i and ℓ on the remaining occupied orbitals.
Viewed as a shift in the binding energy of an electron in orbital ℓ, brought
about by a hole in orbital i, the indirect interactions may be understood
as follows. The passive orbitals relax toward the attractive hole in orbital
i. Electrons in these passive orbitals will then produce a different effective
electrostatic potential at orbital ℓ. The difference, which Hedin and
Johansson termed the "polarization potential" V_p, will increase the energy
of the two-hole state by a relaxation energy

$$R(i\ell) = \langle \ell | V_p(i) | \ell \rangle = \langle \ell | \sum_m [V(\ell m)_i - V(\ell m)] | \ell \rangle \quad . \qquad (5)$$

Here $V(\ell m)$ is the operator that describes the two-electron interaction between
orbitals ℓ and m, including both Coulomb and exchange contributions, in the
neutral atom. The corresponding operator for an ion with a hole in the i
orbital is denoted by $V(\ell m)_i$. The sum is taken over all occupied orbitals
m. Thus, for example, if ℓ and m were an s and a p orbital, one term
in the sum would have an expectation value

$$\langle \ell | V(\ell m)_i - V(\ell m) | \ell \rangle = \mathscr{J}(\ell m)_i - \mathscr{J}(\ell m)$$

$$= F^0(\ell m)_i - F^0(\ell m) - \frac{1}{6} [G^1(\ell m)_i - G^1(\ell m)] \quad . \quad (6)$$

Here again the subscript i denotes a hole in orbital i. The intermediate
step was inserted in Eq. (6) to indicate the relationship between direct and
indirect interactions.

After considering the analogous three steps for the jkℓ hole state
and combining the energies for the three steps in each case, we have

$$E(iℓ) = E(i) + E(ℓ) + \mathcal{F}(iℓ) - R(iℓ)$$

$$E(jkℓ) = E(jk) + E(ℓ) + \mathcal{F}(jk;ℓ) - R(jk;ℓ) \quad . \tag{7}$$

The signs of the $\mathcal{F}$ and R terms indicate that hole-hole interactions are
repulsive, whereas relaxation of electrons toward a hole in an inner shell
has an attractive effect on another such hole (or that this relaxation
increases the binding energy of the corresponding electron[5]). From Eqs. (2),
(2a), (3), and (7), the Auger shift can be expressed as

$$\Delta E(ijk(ℓ)) = \mathcal{F}(iℓ) - \mathcal{F}(jk;ℓ) - R(iℓ) + R(jk;ℓ) \quad . \tag{8}$$

It is useful to note at this point the <u>pairwise additivity</u> of the interactions
of the Auger holes j and k with the "spectator" hole ℓ. Since the direct
terms $\mathcal{F}$ are taken here as mean interactions without regard to the details
of multiplet coupling, it follows immediately that

$$\mathcal{F}(jk;ℓ) = \mathcal{F}(jℓ) + \mathcal{F}(kℓ) \quad . \tag{9}$$

For the indirect terms, pairwise additivity requires that

$$R(jk;ℓ) \cong R(jℓ) + R(kℓ) \quad , \tag{10}$$

which would in turn imply

$$\mathcal{F}(ℓm)_{jk} - \mathcal{F}(ℓm) \cong \mathcal{F}(ℓm)_j + \mathcal{F}(ℓm)_k - 2\mathcal{F}(ℓm) \quad . \tag{11}$$

This relation is neither generally nor exactly true, but it should be a good

approximation for core holes in orbitals j and k in heavy atoms. It

requires, in essence, that the removal of an electron from the j or k

orbital could be treated as a first-order perturbation on the total electrostatic

potential experienced by an electron in the ℓ or m orbital (and that two such

perturbations can be added to simulate the effect of creating holes in

orbitals j and k). In heavy atoms the removal of a core electron would

affect the potential at outer orbitals by a fraction of order $1/Z$. Thus

pairwise additivity of the indirect terms should be approximately observed in

heavy atoms, and Eq. (10) should be accurate to within a few percent. Assuming

pairwise additivity of both direct and indirect interactions, Eq. (8) becomes

$$\Delta E(ijk(\ell)) = \mathcal{F}(i\ell) - \mathcal{F}(j\ell) - \mathcal{F}(k\ell) - R(i\ell) + R(j\ell) + R(k\ell) \quad . \quad (12)$$

Up until this point the discussion has been cast in terms of hole-state

interactions. The use of hole state wavefunctions saves no labor when compared

to the "brute-force" total-energy approach, as both require hole-state

SCF calculations. Now that the hole-state interactions have been decomposed

into pairwise interactions, however, it is possible to make approximations for

the $\mathcal{F}$ and R terms that allow $\Delta E(ijk(\ell))$ to be estimated fairly accurately using

only tabulated integrals obtained from SCF calculations on atomic ground states.

These approximations are described below.

The $\mathcal{F}$ terms can be calculated by simply using tabulated ground-state

Coulomb and exchange integrals. This approach was discussed and used successfully

for treating KLL Auger energies earlier.[5]

Before estimating the relaxation terms R we note that Hedin and

Johansson subdivided two-electron relaxation effects involving holes into

inner-shell, intra-shell, and outer-shell interactions. By explicit calcula-
tions on hole states the inner-shell effects were shown to be negligible.
Intra-shell effects tend to be small in comparison with outer-shell terms,
especially for core-level holes in large atoms, for which outer-shell terms
dominate. In the version of the model used below, only outer-shell terms
are considered and the "equivalent-cores" approximation is made. This approach
has proved very successful for estimating hole-state relaxation energies.[3-5]

The equivalent-cores approximation is well-known, in an empirical
form, in x-ray spectroscopy, having been used as early as 1921 by Wenzel[8] in
connection with x-ray satellites. In this form it involves estimating the
energy of an x-ray transition in element Z in the presence of a core-electron
hole by using the measured transition energy in element Z + 1. Thus the
shielding of the outer orbitals by a core hole is taken as equivalent to that
of a positive charge in the nucleus. We have previously used a form of the
equivalent-cores approximation which is based on changes in Coulomb and exchange
integrals between Z and Z + 1 to estimate shifts in the energies of one- and
two-hole states.[3-5] Because the radii of electronic shells in a given atom
increase dramatically with each unit increase in principal quantum number n,
it suffices, for estimating the interaction with a hole in shell n, to approx-
imate the radial wave functions of orbitals with n' > n by those of the
corresponding orbitals in the next higher element. The additional interaction
of these orbitals with holes in shells of n" < n' is then given by

$$R(n''\lambda'',n'\lambda') = \sum_{\substack{n'\lambda', \\ n' > n,n''}} N(n'\lambda')[\mathcal{G}(n''\lambda'',n'\lambda')_{Z+1} - \mathcal{G}(n''\lambda'',n'\lambda')_{Z})] \quad . \quad (13)$$

Here λ is the orbital angular momentum quantum number, $N(n'\lambda')$ is the occupation number of the $n'\lambda'$ subshell, and the sum is taken over all occupied states with $n' > n$.

The above approximations may appear rather crude. They have good physical bases, however, and can be justified by lengthy but straightforward arguments and calculations, some of which have been given previously.[3-5] While SCF hole-state calculations would ultimately be preferable, the model described above should be adequate in most cases to provide a reasonably accurate estimate of Auger satellite shifts.

A few of the most important types of Auger satellites are categorized according to the relative values of the principal quantum numbers and discussed separately below.

Type I: $n(j) = n(k) = n(i) + 1$. This is the most commonly studied type of Auger transition (KLL, LMM, etc.). There are four satellite subcategories, for different values of $n(\ell)$. For $n(\ell) < n(i)$ the satellite would rarely be observable because the $n(\ell)$ hole would tend to have a shorter lifetime than the $n(i)$ hole and to fill first. The $\not{F}(i\ell)$ term would far outweigh the $\not{F}(jk;\ell)$ terms and the satellite would have a large positive shift.

For $n(\ell) = n(i)$ the $\not{F}$ and R terms show a strong tendency to cancel separately thereby precluding any very general comments about the shifts. The size of the shift should therefore be predicted separately for each transition.

The $n(\ell) = n(j) = n(k)$ case has been the most thoroughly studied experimentally. The shift is given by

$$\Delta E(ijk(\ell)) = \not{F}(i\ell) - 2\not{F}(j\ell) - R(i\ell) + 2R(j\ell) \quad , \qquad (14)$$

where we have assumed the j and k orbitals to contribute equally to the
$\mathcal{F}$ and R terms. This is a very good approximation generally: the $\mathcal{F}(j\ell)$ and
$R(j\ell)$ terms tend to depend only on the principal quantum numbers $n(j)$ and $n(\ell)$.
The last terms in Eq. (14) have the forms

$$R(i\ell) = \sum_{\substack{n' > n(i)+1 \\ \lambda'}} N(n'\lambda')[\mathcal{F}(i,n'\lambda')_{Z+1} - \mathcal{F}(i,n'\lambda')_Z] \quad , \quad (15a)$$

$$R(j\ell) = \sum_{\substack{n' > n(i)+1 \\ \lambda'}} N(n'\lambda')[\mathcal{F}(j,n'\lambda')_{Z+1} - \mathcal{F}(j,n'\lambda')_Z] \quad . \quad (15b)$$

In heavy atoms $\mathcal{F}(i\ell)$ is slightly larger than $\mathcal{F}(j\ell)$ while $R(i\ell)$ and $R(j\ell)$
tend to be about equal in size and half as large as $\mathcal{F}(j\ell)$. Thus $\Delta E(ijk(\ell))$
is always predicted to be negative, as observed.[1,2]

When $n(\ell) > n(j)$ the above discussion still applies, but all the terms
are smaller in magnitude. The shift is still negative, but smaller, and it
decreases with increasing $n(\ell)$. For multiple vacancies in outer shells the
shifts are essentially additive. This is simply another manifestation of
pairwise additivity of hole-state shifts, discussed earlier. Two vacancies
with $n(\ell) = n(j) + 1$ would create a shift about equal to that of a single
$n(\ell) = n(j)$ vacancy, while a single $n(\ell) = n(j) + 1$ vacancy would cause a
shift of about half this size.

Type II: $n(k) > n(i) + 1 \leqslant n(j)$. These are Auger transitions for
which one or both of the final-state holes has a principal quantum number
more than one unit larger than that of the initial state; e.g., and LMN, LMX,
or LNN transition. For brevity only transitions in which $n(\ell) = n(i) + 1$
will be discussed. The shift is now given by Eq. (12).

In the case $n(k) = n(j) + 1 = n(i) + 2$ the magnitudes of the interaction
terms will decrease in the order $\mathcal{F}(i\ell) > \mathcal{F}(j\ell) > \mathcal{F}(k\ell)$ and $R(i\ell) > R(j\ell) > R(k\ell)$.
The combination of terms $\mathcal{F}(i\ell) - \mathcal{F}(j\ell) - \mathcal{F}(k\ell)$ will be small and negative,
while $-R(i\ell) + R(j\ell) + R(k\ell)$ is small and positive. The net near cancellation
leads to a small satellite shift. For the case $n(k) = n(j) = n(i) + 2$ the $\mathcal{F}(i\ell)$
term outweighs both $\mathcal{F}(j\ell)$ and $\mathcal{F}(k\ell)$ and the net shift is large and positive.
The pairwise additivity of shifts in terms of final-state holes is nicely
illustrated by these two cases. Consider as a specific example the shifts
caused by an m vacancy. The LMM transition satellites will show large
negative shifts, while LMN satellite shifts will be small, and LNN shifts
large and positive. These cases are all discussed in the next section.

Other types of satellites should be observable. The above discussion
covers all the cases observed until now. Application of this model to new
cases should be straightforward.

III. NUMERICAL RESULTS

In this section the shifts predicted for $L_3 M_{45} M_{45} (M_{45})$ transitions are presented for elements with atomic number $70 \leqslant Z \leqslant 100$. Also presented are various shifts for bismuth, the most-studied element.[1] Comparisons with experiment are made where possible.

For $L_3 M_{45} M_{45} (M_{45})$ transitions Eq. (14) takes the specific form

$$\Delta E(L_3 M_{45} M_{45} (M_{45})) = \mathscr{F}(2p\ 3d) - 2\mathscr{F}(3d\ 3d) - R(2p\ 3d) + 2R(3d\ 3d) \quad . \quad (16)$$

Standard multiplet theory[7] gives

$$\mathscr{F}(2p\ 3d) = F^0(2p\ 3d) - (1/15)\ G^1(2p\ 3d) - (3/70)\ G^3(2p\ 3d)$$

$$\mathscr{F}(3d\ 3d) = F^0(3d\ 3d) - (2/63)\ F^2(3d\ 3d) - (2/63)\ F^4(3d\ 3d) \quad . \quad (17)$$

Similar expressions can be written for the terms $\mathscr{F}(2p, n'\lambda)$ and $\mathscr{F}(3d, n'\lambda)$ that make up the relaxation terms

$$R(2p\ 3d) = \sum_{n' > 3} N(n'\lambda)[\ \mathscr{F}(2p, n'\lambda)_{Z+1} - \mathscr{F}(2p, n'\lambda)_Z]$$

$$R(3d\ 3d) = \sum_{n' > 3} N(n'\lambda)[\ \mathscr{F}(3d, n'\lambda)_{Z+1} - \mathscr{F}(3d, n'\lambda)_Z] \quad . \quad (18)$$

Numerical values of $\Delta E(L_3 M_{45} M_{45} (M_{45}))$ were calculated from Eqs. (16) - (18) using Mann's[9] values of the Slater integrals. In calculating the relaxation terms it was necessary to use smoothed values in some cases because of changes in open-shell configurations from one element to the next (Z to $Z + 1$). Results

are given in Table I for intervals of $\Delta Z = 5$. The shifts increased regularly

by a factor of 2.5 from $Z = 70$ to $Z = 100$, although the estimated values of

R varied erratically near $Z = 90$. The calculated values of $\Delta E(L_3 M_{45} M_{45}(M_{45}))$

show excellent agreement with experiment in the two elements for which measure-

ments are available: -38 eV vs -36 ± 5 eV[1] in bismuth and -61 eV vs -56 eV[2]

in americium.

Haynes, et al. also reported satellite shifts in bismuth of

$$\Delta E(L_3 MN(M)) \approx 0$$

$$\Delta E(L_3 N_2 N_5(M)) \cong +50 \text{ eV}$$

$$\Delta E(L_3 N_5 N_5(M)) \cong +68 \text{ eV} \quad ,$$

all with "quite large error", for transitions of Type II in the previous

section. The model described herein was used to calculate values of +21,

+76, and +80 eV, respectively, for these three shifts. Agreement with experi-

ment is apparently satisfactory in view of the large errors indicated by

Haynes et al. Clearly this comparison does not provide a very sensitive test,

but the trend from negative shifts in LMM to small shifts in LMN to large

positive shifts in LNN transitions shows encouragingly good agreement of this

model with experiment.

IV. SUMMARY

The origins of Auger satellite energy shifts were categorized into

direct and indirect (or relaxation) interactions. They were further decomposed,

within a self-consistent-field framework, into their component two-electron

interaction terms. The pairwise additivity of shifts was thus made

apparent: the energy shift of a two-hole state due to a "spectator" vacancy

is essentially the sum of the shifts that it induces in each of the corresponding

one-hole states, while the effect of two spectator vacancies on the energy of

a hole state is the sum of their individual effects. These results can be

used for predicting satellite shifts. Thus, for example, we would expect

$$\Delta E(LMN(M)) \cong \frac{1}{2} \left[\Delta E(LMM(M)) + \Delta E(LNN(M)) \right]$$

$$\Delta E(LMM(N^2)) \cong 2\Delta E(LMM(N)) \quad ,$$

etc.

Direct-interaction terms can be calculated in a straightforward way,

using multiplet coupling theory and Mann's two-electron integrals. A rigorous

calculation of the indirect, or relaxation, terms would require hole-state SCF

results. These terms can be estimated to a good approximation, however, by

using an equivalent-cores model together with ground-state two-electron integrals.

Numerical estimates of Auger satellite shifts for the $(L_3 M_{45} M_{45} (M_{45}))$ transi-

tion show ΔE increasing in magnitude by a factor of 2.5 from Yb (Z = 70) to

Fm (Z = 100). Excellent agreement with experiment is obtained for Bi (Z = 85)

and Am (Z = 95). Satisfactory agreement is obtained for other, less-accurately

known satellite shifts.

FOOTNOTES AND REFERENCES

*Work performed under the auspices of the U. S. Atomic Energy Commission.

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Table I. $L_3M_{45}M_{45}(M_{45})$ Auger Satellite Energy Shifts (in Rydbergs)

Element	Z	$\mathscr{F}$(2p 3d)	$\mathscr{F}$(3d 3d)	R(2p 3d)	R(3d 3d)	$\Delta E(L_3M_{45}M_{45}(M_{45}))$	ΔE_{expt}
Yb	70	11.45	8.94	4.96	4.73	−1.93(−26 eV)	
Re	75	12.46	9.75	5.41	5.18	−2.14(−29 eV)	
Hg	80	13.47	10.56	5.87	5.64	−2.24(−30.5 eV)	
Bi	85	14.47	11.37	5.93	5.70	−2.80(−38 eV)	−36(5) eV[a]
Th	90	15.48	12.19	5.22	4.99	−4.14(−56 eV)	
Am	95	16.49	13.00	5.60	5.30	−4.51(−61 eV)	−56 eV[b]
Fm	100	17.50	13.82	5.98	5.68	−4.76(−65 eV)	

[a]Ref. 1.

[b]Ref. 2.

FIGURE CAPTION

Fig. 1. Energy-level diagram illustrating the formation of the "spectator"

hole states $i\ell$ and $jk\ell$ from the "regular" hole states i and jk, by a

hypothetical three-step process. Corresponding Auger transitions are shown.

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Fig. 1