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Chemical-bond dependence of Coulomb capture of pions in transition-metal borides

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Atomic Coulomb capture ratios $A(\mathrm{B},M)$ for pions have been measured in chromium, iron, and nickel borides. Those for mixtures of respective metals and boron were also obtained. The ratios for the compounds are less than half of those of the mixtures and depend on the species of the borides. These results for the borides are not predicted by any model. The mesomolecular model revised to take account of the Auger process reproduces the present results. The electron charge densities deduced by applying the model are compared with theoretical predictions.

I. INTRODUCTION

Negatively charged particles, pions and muons, which are slowed down in a chemical compound will be captured in one of the constituents. The periodic behavior of these atomic (per atom) Coulomb capture ratios (ACR), which was first reported by Zinov *et al.*,[1] has been very well studied experimentally for oxides and halides (cf. Ref. 2 and literature cited therein). Theories have been developed which reproduce this periodic behavior by relating it to the atomic radius[3] or to the number of weakly bound electrons.[4,5] Only very recently, however, it was found that the molecular structure or solid-state structure also influences the ACR.[6–9] This fact indicates that mesons might be used as a probe to measure the spatial distribution of bonding electrons. The mesomolecular model[4] developed by Schneuwly *et al.* should be tested from this point of view.

In our previous report,[9] we studied the ACR for compounds of very light elements, beryllium and boron, where bonding electrons were expected to exert a strong influence, and very distinct molecular effects were found for the compounds. In the present study the measurements of the ACR are extended to several transition-metal borides. Metallic partners are the transition elements of chromium, iron, and nickel. It is widely accepted that essentially the electrons bound with less than several tens of electron volts are responsible for capturing pions or muons.[4,5] In chromium, iron, and nickel, therefore, twenty electrons, at most, would contribute in the process. Then, the ACR could be affected strongly by the spatial distribution of the electrons in the outermost $3d$ and $4s$ electron shells, which depends on the molecular species of the metal borides.

The mesomolecular model developed by Schneuwly *et al.*[4] was applied to the present cases, and revised further to reproduce the experimental data. The electron densities deduced with the revised mesomolecular model are compared with theoretical predictions.[10–15]

II. EXPERIMENTAL PROCEDURE

A. Experimental arrangement

The experiments were carried out at a $\pi\mu$ channel of the National Laboratory for High Energy Physics (KEK) with 140 MeV/c negative pions (momentum bite $\Delta p/p$ of $\pm 6\%$ and a beam duty factor of 20% at a frequency of 0.4 Hz). The experimental arrangement is shown in Fig. 1. Pions, focused onto a target, were degraded with a 7.7-cm-thick carbon plate and collimated with a 1-cm-thick iron plate with a 4×4 cm^2 hole. Pions stopped in

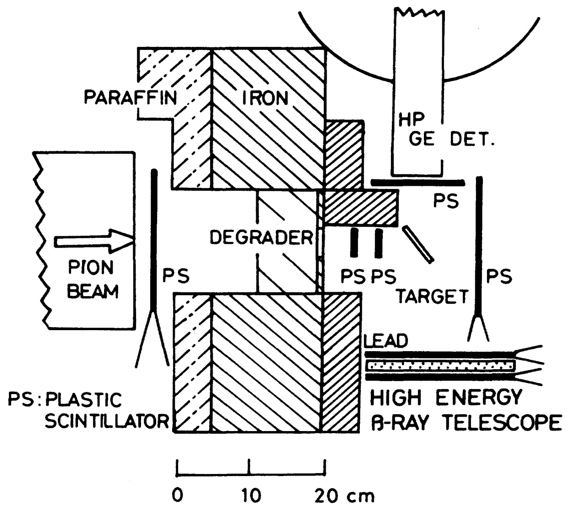

FIG. 1. Schematic drawing of experimental setup.

the target were counted with a conventional counter telescope composed of one 20×20 cm^2, two 4×4 cm^2, and one 20×20 cm^2 (veto) thin plastic scintillators. Targets were metal boride compounds of Cr_2B, CrB, CrB_2, Fe_2B, FeB, Ni_3B, Ni_2B, Ni_3B_2, and NiB, and mixtures of these elements with the same atomic fractions as in the compounds. The metal borides were purchased from Cerac Inc.[16] and were typically 99% pure. Mixtures were prepared by uniformly mixing metal powder (typically 4—9-μm-diam particles) with amorphous boron powder (0.8—1.2-μm-diam particles). For checking the effect of particle size on the ACR different sizes of metal powder were used. Powder samples were contained in rectangular-shaped target holders composed of styrofoam frames with an interior cross section of 4×8 cm^2 and with top and bottom windows of 100-μm-thick Mylar film. All targets were 1.75 g/cm^2 in areal thickness and were positioned 10 cm behind the collimator at an angle of 45° to the beam direction.

The pionic x rays were measured with an Ortec intrinsic pure germanium detector for low-energy photon spectrometry (LEPS) 2.5 cm in diameter and 0.5 cm thick. The energy resolution of the detector was 0.4 keV for 5.9-keV photons. The detector was set 12 cm from the target and was shielded by paraffin, iron, and lead collimators to reduce the background from photons, neutrons, and scattered charged particles. The pionic x rays, detected with the LEPS operated in coincidence with stopped-pion events, were analyzed with an Ortec 800 analog-to-digital converter (ADC) and recorded onto a floppy disk for subsequent spectrum analysis. The detector-efficiency curve was obtained with a mixed standard of ^{60}Co, ^{137}Cs, and ^{241}Am activities of known intensities and also with ^{152}Eu of known activity. Each of the two standard sources was mixed homogeneously in FeB powder and counted in the styrofoam container, thereby correcting for self-absorption of photons with energy greater than or equal to 50 keV. The efficiency of the fast coincidence circuit was determined to be 0.63±0.02 by comparing the coincidence events with singles events for the pionic 4⟶3 x rays from pure-iron samples.

In a typical run, 7.5×10^4 pions and 1.4×10^4 muons per accelerator beam pulse entered the target, and 3×10^4 pions stopped in every pulse. Each spectrum was accumulated for 4 to 8 h. Delayed coincidence spectra and singles spectra were taken to identify random coincidence events.

The intensity of the 65-keV 2⟶1 x rays from the pionic boron atom was too weak to be accurately detected. As an alternative method for measuring the capture rates of pions in boron, β rays ($E_{\beta\max}=12.96$ MeV) from ^8Li ($T_{1/2}=838$ ms),[17] produced after the pion absorption in $^{10,11}B$ ($^{10,11}B+\pi^-\longrightarrow{}^8Li+p+xn$) were detected with a high-energy counter telescope[18] composed of two plastic scintillators and an aluminum absorber sandwiched between them, as shown in Fig. 1.

III. RESULTS

A. Pionic x-ray spectra

The pionic x-ray spectra were observed for the compounds and the mixtures. Spectra were also taken for the pure elements, chromium, iron, and nickel. Typical spectra are shown in Fig. 2 for the Cr_2B compound. The pionic x-ray peaks of the transitions to principal quantum numbers $n_0=2$, 3, 4, and 5 are observed. Other peaks were attributed to γ rays emitted after the pion absorption in a chromium nucleus. Data for a blank sample holder, a carbon plate, and water were used for the critical evaluation of background subtraction. The 3⟶2 peak was broadened by the competing nuclear absorption, and its energy increased by 4.1, 5.7, and 7.9 keV for Cr, Fe, and Ni, respectively, compared with calculated values without the nuclear absorption. The broadening and energy shift were not observed for transitions to states with $n_0\geq3$ in the present experimental conditions.

In Fig. 3, transition intensities for the pionic chromium, iron, and nickel atoms were plotted versus the difference (Δn) of the principal quantum numbers between the states for the series to $n_0=3$, 4, and 5. No molecular dependence of x-ray intensity pattern was observed among the compounds, the mixtures, and the pure metals. Naumann *et al.*[19] also claimed that the muonic *K* x-ray intensity patterns measured for $FeCl_3$, Fe_2O_3, and $Fe(NO_3)_3$ were indistinguishable. Hild *et al.*[20] found no difference between CrO_3 and Cr_2O_3. Therefore, averaged values among the samples were plotted in Fig. 3. Gross dependences of the transition intensity I on Δn are $I\propto\Delta n^{-2.4}$, $I\propto\Delta n^{-2.1}$, and $I\propto\Delta n^{-2.1}$ for $n_0=3$, 4, and 5 series in chromium, iron, and nickel, except for the $n_0=4$ series in

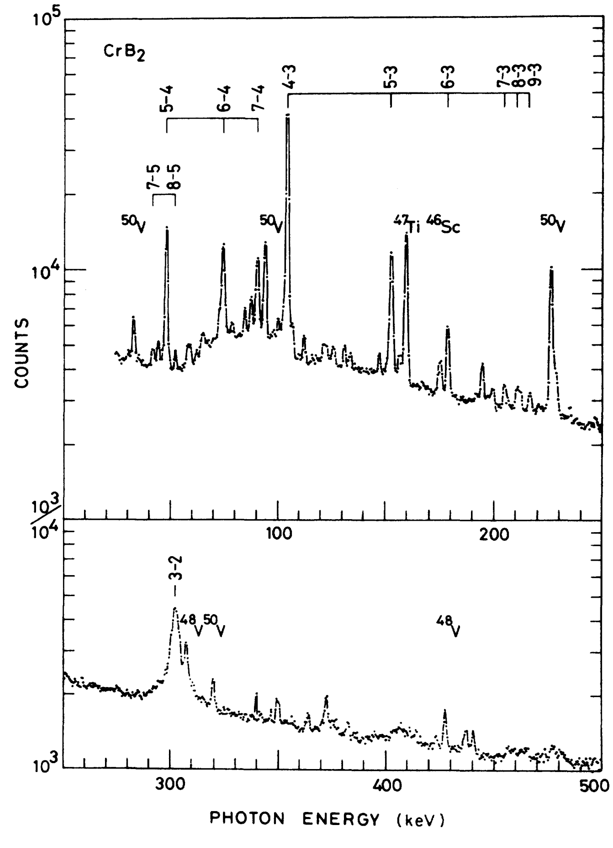

FIG. 2. Pionic x-ray spectrum for CrB_2.

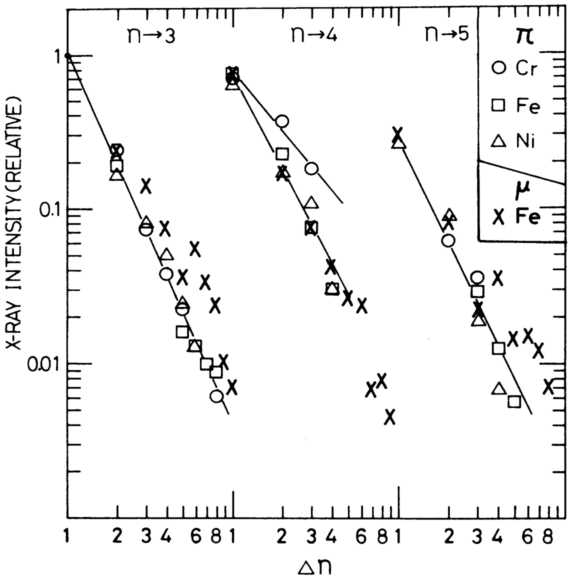

FIG. 3. Pionic x-ray intensities in chromium, iron, and nickel. Intensities are average values among compounds, mixtures, and pure metals. Crosses are muonic x-ray intensities from Ref. 21.

chromium where $I \propto \Delta n^{-1/3}$ dependence was found. The x-ray intensities for muonic iron atoms[21] are also shown in Fig. 3. The results of the pionic and muonic irons show that the transitions from higher states were populated more in the muonic atoms than in pionic atoms, which indicates that qualitatively the initial angular momentum is higher for the pionic atoms than for the muonic atoms.

B. Pionic x-ray yields per pion stop

Pionic x-ray yields per pion stop were obtained for the 4→3 peak in the spectra of the separate elements of chromium, iron, and nickel. Here, the peak areas in the spectra were corrected for detection efficiencies and the fast coincidence efficiency. The number of stopped pions was obtained by correcting the total events for the contribution of the muons stopped (16%) in the targets. The results are shown in Table I along with previous data.[22,23] Our data agree with the mean values of the results of Refs. 22 and 23 within the quoted errors.

C. Decays of β rays from ^{8}Li

Pions captured by boron atoms are finally absorbed by the nucleus and the emission of protons and neutrons will follow. Main radioactive products are expected to be ^{6}He ($T_{1/2}=806.7$ ms, $E_{\beta\max}=3.5$ MeV), ^{8}He (119 ms, 10.6 MeV), ^{8}Li (838 ms, 12.96 MeV), and ^{9}Li (178.3 ms, 13.6 MeV).[17] Here, ^{8}Li was discriminated from low-energy β-ray emitters using the high-energy β-ray telescope.[18] In addition, the proton beams were extracted from the 12-GeV proton synchrotron for 0.53 s every 2.58 s. Therefore, β rays were measured in coincidence with the beam-on periods in multiscaling mode. The activity of ^{8}Li was thus obtained from the decay curve analysis of the β-ray measurement shown in Fig. 4.

TABLE I. Intensities (I) of pionic x rays per stopped pion.

Element	Transition	I (%) This work	Other work	
B	2→1	8.6±0.6		
Cr	4→3	23.1±1.6	23.5±2.2[a]	26.6±1.9[b]
Fe	4→3	28.8±2.0	23.7±2.2[a]	30.2±1.9[b]
Ni	4→3	29.9±2.1	27.7±2.9[a]	34.1±1.9[b]

[a] Reference 22.
[b] Reference 23.

D. Atomic Coulomb capture ratio

Values of the atomic Coulomb capture ratio of boron to metal $A(\mathrm{B},M)$ were determined the following way. First, we measured and analyzed the x-ray spectra for thin, boron-rich mixtures (thickness, 0.18 g/cm^2 and atomic ratio of boron to metal, 10/1), where the self-absorption of the 65-keV 2→1 x ray from the pionic boron atom was small. We applied corrections for detection efficiency and x-ray yield per pion stop to the photo peak counts of the relevant transitions shown in Table I. Values of $A(\mathrm{B},M)$ are shown in Table II. Second, the apparent capture ratios for the mixtures obtained from the measurements of

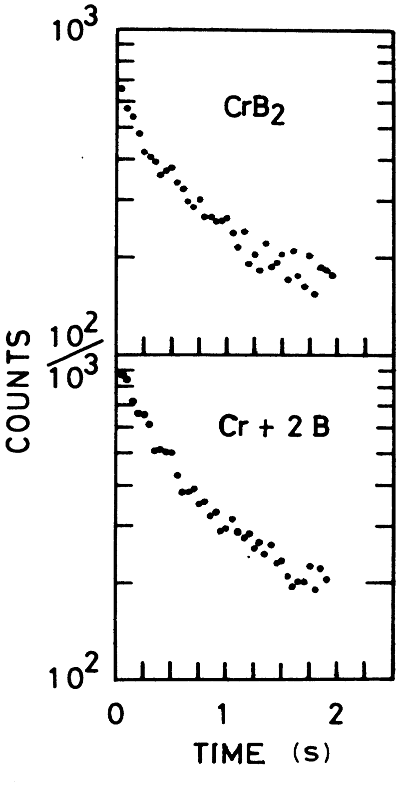

FIG. 4. Decay of β rays emitted from ^{8}Li and ^{9}Li formed after pion absorption of ^{10,11}B in CrB$_2$.

TABLE II. Experimental atomic Coulomb capture ratios $A(\mathrm{B},M)$ for compounds and mixtures. Predictions are also presented.

Compounds and mixtures	$A(\mathrm{B},M)$				
	Experiment	Calculated			
Mixture (Cr + 10B)	0.32±0.02	0.38^{a}			
Cr_2B	0.08±0.01		0.27^{b}	0.22^{c}	0.043^{d}
CrB	0.12±0.01				
CrB_2	0.14±0.01				
Mixture (Fe + 10B)	0.35±0.03	0.36^{a}			
Fe_2B	0.09±0.02		0.25^{b}	0.20^{c}	0.037^{d}
FeB	0.11±0.02				
Mixture (Ni + 10B)	0.35±0.02	0.35^{a}			
Ni_3B	0.09±0.02		0.23^{b}	0.19^{c}	0.034^{d}
Ni_2B	0.10±0.02				
Ni_3B_2	0.10±0.02				
NiB	0.12±0.01				

[a] Reference 26.
[b] Reference 3.
[c] Reference 5.
[d] Reference 4.

x-ray spectra for metal constituents and β-ray decay for boron were normalized to the values of $A(\mathrm{B},M)$ for the 10:1 atomic fraction mixtures because it had been confirmed that the ACR is independent of the atomic fraction for condensed mixtures.[24,25] Finally, the values of $A(\mathrm{B},M)$ for the compounds were obtained relative to the ones for the corresponding mixtures with the same constituents. The results are shown in Table II along with the predicted values.[3,4,5,26]

IV. DISCUSSION

A. Atomic Coulomb capture ratios

As shown in Table II and Fig. 5, when elements form a molecular compound, $A(\mathrm{B},M)$ changes substantially from the value for the mixtures. The fact is not attributed to the particle-sizes effect on the ACR for the mixtures, because the effect was checked carefully with two different particle sizes of metal powder (4—9-μm-diam and 50-μm-diam particles), but no effect was found outside of the statistical uncertainty. Similar molecular effects have already been observed by Knight *et al.*[6] and by our previous experiment.[9] Another finding which was pointed out in Ref. 9 has been confirmed by the present experiments, that is, the ACR depends on the atomic fraction of the constituent elements in the molecule or on the structure of the molecule.

Theoretical predictions by Vasilyev *et al.*,[26] which do not show any periodicity, reproduce the experimental results for the mixtures. A semiclassical model by Daniel[3] and a quantum-mechanical description by von Egidy *et al.*[5] give values lying between those of the mixtures and the compounds.

So far only the mesomolecular model by Schneuwly *et al.*[4] explicitly includes the chemical dependence of the ACR, but fails to reproduce the present experimental results. The predicted values are much lower than the experimental data for the chemical compounds. Here, the ionicity σ of the chemical bond was estimated from Pauling's electron negativity. However, the ionicity may change depending on the molecular structure. Therefore, we took the ionicity σ as an adjustable parameter which produced the best agreement between the model predic-

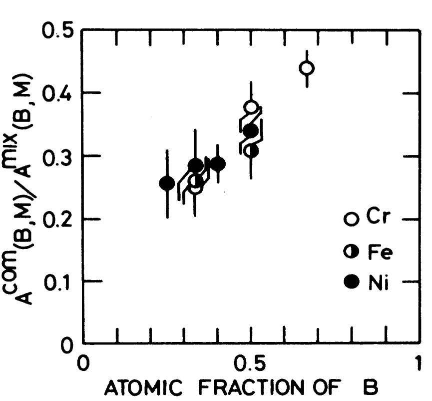

FIG. 5. Ratios of experimental atomic Coulomb capture ratios between metal borides and mixtures as a function of atomic fraction of boron.

tions and the experimental values. This procedure was used in Refs. 8 and 9. Other parameters were fixed to the values reported in Ref. 4. The resulting values of the ionicity parameter σ are given in Table III. The deduced ionicity parameters, however, give unacceptable atomic charges for the constituents in the metal borides.

B. Modified mesomolecular model

The mesomolecular model was introduced by Ponomarev[27] to explain the capture probability of pions in hydrogen in hydrogen-containing compounds. Schneuwly *et al.*[4] extended it to general compounds of the composition $(Z_1)_k(Z_2)_l$. In the model, the process of the capture of a "meson" is divided into two steps. First, the meson is trapped in an atomic or molecular orbit with low binding energy after knocking-out an electron. Then, the meson trapped in the mesomolecular state deexcites to an atomic state through radiative transitions.

We follow basic ideas of the mesomolecular model, but we attempt to modify the cascade process of the molecular to atomic states. That is, the mesomolecular and electronic orbitals are located closely to one another. Then, the interaction between the meson and the electrons should be strong and a high probability of Auger process is expected. The "redistribution factor" q_1 from the molecular state to the atomic states of the constituent Z_1 is then simply described for the molecule $(Z_1)_k(Z_2)_l$ with $Z_1 < Z_2$ by

$$q_1 = [1 + (Z_{\text{eff2}}/Z_{\text{eff1}})]^{-1} , \tag{1}$$

where the Z_{eff}'s are the effective electron numbers of atoms Z_1 and Z_2, which are assumed to be identical to the number of electrons participating in the first step of trapping the meson.

Thus, the atomic Coulomb capture ratio $A(Z_1,Z_2)$ is redefined as

$$A(Z_1,Z_2) = \frac{l}{k}\,\frac{kn_1 + v\omega_1}{ln_2 + v(1-\omega_1)} , \tag{2}$$

where ω_1 is a probability that a meson on the molecular orbital deexcites into mesoatomic orbitals of atom Z_1. Here ω_1 is assumed to be given by the following formula for the long-lived mesomolecular state in Ref. 4 with the "localization factor" p_1, which is expressed as $p_1 = \frac{1}{2}(1+\sigma)$:

$$\omega_1 = \frac{p_1 q_1}{[p_1 q_1 + (1-p_1)(1-q_1)]} , \tag{3}$$

$$\omega_1 = \frac{Z_{\text{eff1}}(1+\sigma)}{Z_{\text{eff1}}(1+\sigma) + Z_{\text{eff2}}(1-\sigma)} , \tag{4}$$

and v is the sum of valence electrons v_1 and v_2 of respective atoms Z_1 and Z_2, n is the number of core electrons with the binding energy E less than a cutoff energy E_0, and $Z_{\text{eff}i} = n_i + v_i$. For elements with Z above 10, E_0 fixed to about 100 keV, and for the lighter elements, E_0 is slightly adjusted element by element.

TABLE III. Values of the ionicity parameter σ_m in the model by Schneuwly *et al.* (Ref. 4) required to achieve the best agreement between predicted and experimental atomic Coulomb capture ratios $A(Z_1,Z_2)$ with $Z_1 < Z_2$. v is the sum of valence electrons in B and metal (Cr, Fe, or Ni). n is the number of core electrons with binding energy less than a cutoff energy, E_0 ($\sim$100 keV).

Compound	Parameters			
	v	n_1	n_2	σ_m
Cr_2B	15	0	6.16	0.27
CrB	9	0	6.16	0.67
CrB_2	12	0	6.16	0.84
Fe_2B	19	0	5.06	0.28
FeB	11	0	5.06	0.64
Ni_3B	33	0	4.20	0.15
Ni_2B	23	0	4.20	0.36
Ni_3B_2	36	0	4.20	0.49
NiB	13	0	4.20	0.67

C. Atomic charges deduced from the modified mesomolecular model

The modified mesomolecular model was checked with the data on oxides and fluorides of chromium, iron, and nickel. Measured results are shown in Table IV along with adopted parameters. The results are very encouraging and the resultant atomic charges are acceptable in comparison with the molecular structure generally understood for these compounds.

Next, we find the parameters for the effective number of electrons for metals and applied them to the metal borides. Results are presented in Table IV. For every metal boride shown in Table IV, electron density increases from metal to boron with increasing an atomic fraction of boron.

Recently considerable attention has been given to metal borides and sophisticated theoretical calculations have been performed to obtain information about the electronic band structures and densities of states. Armstrong performed calculations with a linear combination of atomic orbitals (LCAO) method and obtained charge distributions from the density of states for CrB_2 (Ref. 10) and FeB (Ref. 11). The results are shown in Table IV. Samsanov *et al.*[12] obtained the same direction of the electron drift with similar calculations. Electrons transfer from metal to boron for both borides, which agrees qualitatively with the present results. An augmented-plane-wave band calculation[13] for CrB_2 gave similar results, but the Korringa-Kohn-Rostoker method[14] gave a contrary result. Another band calculation[15] gave an electron movement from metal to boron for FeB, but no evidence for donation by boron of electrons to the *d* band for Fe_2B.

V. SUMMARY

Our principal results and conclusions from this investigation are as follows.

(i) The values of $A(\mathrm{B},M)$ for the metal borides are less than half of those for the mixtures. This fact shows the influence of the molecular structure.

(ii) Furthermore, the value of $A(\mathrm{B},M)$ depends on the

TABLE IV. Values of ionicity parameter σ_m and atomic charges deduced from a modified mesomolecular model.

								Atomic charge			
		Parameters						This work		Prediction	
Compound	$A(Z_1,Z_2)$[a]	v	n_1	n_2	$Z_{\text{eff}1}$	$Z_{\text{eff}2}$	σ_m	Z_1	Z_2	Z_1	Z_2
Cr_2B	0.08±0.01	15	1	8	4	14	−0.90	2.2	−1.1		
CrB	0.12±0.01	9	1	8	4	14	−0.43	−0.42	0.42		
CrB_2	0.14±0.01	12	1	8	4	14	0.03	−0.10	0.20	−0.76	1.52[b]
Fe_2B	0.09±0.02	19	1	8	4	16	−0.82	1.3	−0.65		
FeB	0.11±0.02	11	1	8	4	16	−0.44	−0.10	0.10	−0.31	0.31[c]
Ni_3B	0.09±0.02	33	1	6	4	16	−0.88	1.0	−0.33		
Ni_2B	0.10±0.02	23	1	6	4	16	−0.78	−0.47	0.24		
Ni_3B_2	0.10±0.02	36	1	6	4	16	−0.70	−0.31	0.21		
NiB	0.12±0.01	13	1	6	4	16	−0.44	−0.62	0.62		
Cr_2O_3	0.33±0.01[d]	30	0	8	6	14	0.60	−2.0	3.0		
CrO_3	0.31±0.03[e]	24	0	8	6	14	0.95	−1.8	5.4		
Fe_2O_3	0.31±0.02[f]	34	0	8	6	16	0.40	−2.0	3.0		
Ni_2O_3	0.38±0.03[f]	38	0	6	6	16	0.40	−2.9	4.4		
FeF_3	0.41±0.07[f]	29	0	8	7	16	0.69	−1.2	3.5		
NiF_2	0.32±0.02[f]	24	0	6	7	16	0.37	−1.2	2.4		

[a] $Z_1 < Z_2$.
[b] Reference 10.
[c] Reference 11.
[d] A mean value between ones given in References 2 and 20.
[e] Reference 20.
[f] Reference 2 and literature cited therein.

composition of the borides, increasing slightly with increase in the atomic fraction of boron.

(iii) Previous models, including the mesomolecular model by Schneuwly *et al.*, cannot reproduce the present results for the metal borides.

(iv) The mesomolecular model revised to account for the Auger process, gives a reasonably good comparison with our experimental results.

(v) The deduced electron charge densities are compared with theoretical predictions, and qualitatively agree with them.

(vi) Dependence of $A(\mathrm{B},M)$ in mixtures on particle size was not observed.

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