and 1.13 mm/s and ΔE_Q values of 0.50 and 2.69 mm/s, respectively. In addition, the Mössbauer spectrum of **2** shows only one quadrupole doublet (δ, 1.20 mm/s; ΔE_Q, 2.72 mm/s) associated with the high spin ferrous centers. The magnetic moments of **1** and **2** at 200 K are 6.62 and 6.88 μB/complex, respectively. Complex **1** does not exhibit any EPR features at $g < 2$, a result that is similar to that found for the previously reported mixed-valence complex, $[Fe_2HXTA(OAc)_2]^{2-}$ [5] and the reduced uteroferrin–phosphate complex.[23] Complex **2**, on the other hand, exhibits a broad EPR signal at low field with maximum amplitude at 500 G.[24] This is reminiscent of the signal found for deoxyhemerythrin azide, which is proposed to arise from a $\Delta M_s = 8$ transition.[25] Detailed magnetic and spectroscopic studies of these complexes are in progress.

In summary, **1** and **2** represent the first structurally characterized $Fe^{II}Fe^{III}$ and $Fe^{II}Fe^{II}$ redox pair that contain the biologically important triply bridged diiron core. The two complexes have significant differences in their structural and physical properties. Furthermore, the dependence of the physical properties on the bridging and terminal ligands in these complexes should provide important insights into the magnetic and electronic interactions of such binuclear iron centers in biology.

Acknowledgment. This work was supported by the National Institutes of Health Grant GM-38767. A.S.B. is grateful for a N.I.H. Postdoctoral Fellowship (GM-11533). We thank Dr. Vasilios Papaefthymiou for the Mössbauer measurements, Dr. Edmund Day and Jacques Bonvoisin for the magnetic measurements, and Professor Doyle Britton and Paul Boyle for their efforts on the X-ray crystal structure determinations.

Supplementary Material Available: Tables of atomic positional and thermal parameters for $[Fe^{II}Fe^{III}BPMP(OPr)_2](BPh_4)_2 \cdot CH_3COCH_3$ and $[Fe^{II}Fe^{II}BPMP(OPr)_2](BPh_4) \cdot 0.8CH_2Cl_2$ (20 pages). Ordering information is given on any current masthead page.

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Alteration of Bleomycin Cleavage Specificity in a Platinated DNA Oligomer of Defined Structure

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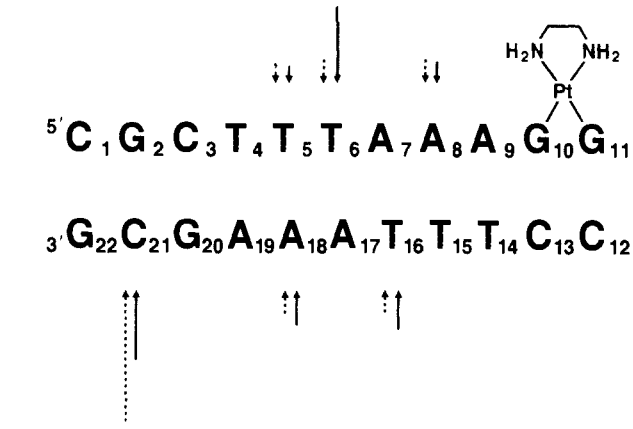
The bleomycins (BLMs) are antitumor antibiotics that are thought to act via metal ion and oxygen-dependent oxidative damage to DNA.[2,3] At least for Fe·BLM, available evidence suggests that degradation involves oxygenation of the deoxyribose moiety at C-4′H.[4] Bleomycin-mediated DNA degradation occurs with considerable selectivity for the pyrimidines in a subset of all 5′-GT-3′ and 5′-GC-3′ sequences.[5,6] Interestingly, the extent of cleavage at preferred sites differed among individual bleomycin congeners,[6f,7] including some having a different spectrum of activity

Chart I[a]



[a] Fe·BLM-mediated strand scission of a platinated (↑) and nonplatinated (↟) DNA oligomer. The extent of cleavage at individual positions was proportional to the length of the arrows shown. The extent of DNA cleavage was determined by HPLC analysis and quantification of individual products (Table I and ref 15). In order to identify the site(s) of enhanced Fe(II)·BLM modification in the platinated oligonucleotide, each of the strands in the duplex was separately 5′-^{32}P end labeled and annealed to its complementary unlabeled strand. The four duplexes (platinated and unplatinated, with the individual strands ^{32}P-end labeled) were subjected to Fe(II)·BLM A_2 digestion, and the relative extents of cleavage at individual positions were determined by densitometry of polyacrylamide gels (Table II).[20] The HPLC and sequence analysis data were found to correlate well.

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Table I. Quantitative Analysis of BLM-Mediated Product Formation[a]

5′CGCTTTAAAGG3′ GCGAAATTTCC	products[b] (μM) cytosine + cytosine propenal	thymine + thymine propenal	adenine + adenine propenal	total products[c]
unplatinated	15.1	6.8	7.8	29.7 ± 0.6
platinated[d]	10.4	14.1	7.5	32.1 ± 1.7

[a] Reaction mixtures (50 μL total volume) contained the appropriate oligonucleotide duplex (1 mM final nucleotide concentration) in 50 mM sodium cacodylate (pH 7), which was treated sequentially with BLM A_2 (0.11 mM final concentration) and (0.11 mM) $Fe(NH_4)_2(SO_4)_2$. The combined solution was maintained at 0 °C for 15 min and then analyzed immediately by HPLC as described,[7e,15] by using authentic synthetic samples as standards. [b] Mean from at least three determinations. [c] Total product is equal to the sum of all free bases and base propenals. [d] Platination was carried out in 20 mM $NaClO_4$ (pH 5.5) at 37 °C for 48 h. The desalted, platinated DNA showed a UV characteristic of platinated DNA with a 2-nm red shift to 260 nm, relative to the nonplatinated oligomer.[16] The determination of platination adducts was performed by enzymatic digestion and HPLC analysis as described previously.[12h] In addition to the predominant intrastrand crosslink at $G_{10}G_{11}$, minor and almost equal amounts of 5′-AG and 5′-GNG crosslinks were observed.

in experimental tumor models.[8] Changes in sequence selectivity have also been noted when single BLM derivatives were activated in the presence of different metals[3h-j] and for Fe(II)·BLM A_2 upon DNA methylation.[9]

Bleomycin is commonly used therapeutically in combination with *cis*-diamminedichloroplatinum(II) (*cis*-DDP).[10,11] The latter also acts at the level of DNA via Pt-centered intrastrand crosslinks; the most abundant lesions involve the N-7 positions of adjacent guanosines.[12] It has been observed that platination of DNA caused major alterations in the sequence specificity of Fe·BLM A_2-mediated cleavage, including the appearance of new cleavage sites and the suppression of others.[13] However, since the position and extent of platination in the DNA was neither controlled nor defined, the exact molecular basis for the alteration in DNA cleavage cannot be specified.

To better understand the molecular basis of altered DNA cleavage, we have studied this phenomenon in a well-defined system.[14] By the use of oligomers with a strong BLM cleavage site and a separate preferred platination site we illustrate (1) a dramatic *alteration* in Fe·BLM sequence selectivity when the Pt center and C-4′H ribose of the normally preferred BLM site were separated by seven base pairs, (2) a sharp *decrease* in the overall extent of cleavage when the Pt center and normal cleavage site were four base pairs apart, (3) the generation of a novel preferred sequence for Fe·BLM via DNA platination, and (4) no change in the actual chemistry of oligomer degradation upon platination.

Oligomers $d(CGCT_3A_3G_2)$ and $d(C_2T_3A_3GCG)$ were prepared as described[15] and annealed. It was anticipated that Fe·BLM-mediated modification of this duplex would occur primarily at C_{21}[7e,15] (Chart I) and platination preferably at $G_{10}G_{11}$.[12] In fact, treatment with 1 equiv of hydrated *cis*-dichloro(ethylenediammine)platinum(II) (*cis*-DEP) gave quantitative platination of the oligomer, >90% of which was localized at $G_{10}G_{11}$ (Table I); *cis*-DEP and *cis*-DDP form structurally similar adducts.[12e,g]

Table II. Densitometric Analysis of BLM-Mediated Cleavage of Platinated and Unplatinated $^{5'CGCTTTAAAGG}_{3'GCGAAATTTCC}$[a]

5′CGCTTTAAAGG 3′GCGAAATTTCC	relative band intensities T_5	T_6	A_8	T_{16}	A_{18}	C_{21}
unplatinated	0.14	0.20	0.24	0.20	0.20	1.00
platinated	0.12	0.60	0.14	0.30	0.27	0.53

[a] Relative intensities of the bands were measured and normalized to correct for the amount of substrate DNA in each gel lane; the numbers are expressed relative to an arbitrarily chosen value of 1.00 for C_{21}.

As expected based on work with 5′-$d(CGCT_3A_3CGC)$,[15] treatment of the unplatinated undecanucleotide with Fe(II)·BLM A_2 (1.2 equiv activated BLM[15]/duplex) resulted in modification primarily at C_{21} (Table I, Chart I) as judged by HPLC analysis in comparison with synthetic standards.[15] Analogous cleavage of the platinated oligomer afforded essentially the same total amount of products, but the *ratio* of products formed from C and T differed dramatically (2.2 versus 0.7; Table I).

Because total product formation for both duplexes was less than the amount of activated Fe·BLM,[15] we suggest that the similarity in total product yield resulted from a conformationally induced alteration of oligonucleotide structure that both enhanced the efficiency of cleavage at one or more T residues and decreased the efficiency of cleavage at C_{21}.[17] Consistent with this interpretation were the observations that platination lowered the melting temperature of the oligomer (37 °C → 27 °C) and effected a small change in the shape of the CD spectrum, which was also shifted to longer wavelength (271 → 276 nm and 244 → 246 nm at the maximum and minimum values of θ). Further support for this interpretation derived from the observation that BLM-mediated products from isomeric oligomer $^{5'CGCTTTGGAAA}_{3'GCGAAACCTTT}$ diminished (29.5 μM → 17.3 μM) upon analogous platination of G_7G_8. Here, platination resulted in diminution of cleavage at C (16.8 μM → 10.9 μM) but also in a *decrease* in cleavage at T (6.2 μM → 3.4 μM) (cf. Table I). In the context of interpreting the observed changes in specificity, it is of interest that NMR studies indicate that the platinated G residues remain H-bonded to their complementary C's at room temperature.[16,18] There is,

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however, severe distortion of the helix;[12,19] consistent with our data (vide supra), DNA melting curves[18] show the relative lability of the duplex structure.

Gel sequence analysis[20] of the platinated oligomer (Table II; Chart I) revealed a threefold increase in BLM-mediated damage at T_6, which became the primary site of damage; a significant increase in cleavage at T_{16} was also noted. The emergence of TT as a preferred Fe(II)·BLM cleavage site is unprecedented and provides an opportunity to identify those features in DNA that render the duplex susceptible to Fe·BLM-mediated modification.

The observed changes in sequence specificity are entirely consistent with those reported earlier with the use of *cis*-DDP and much longer DNA fragments.[13] However, this study extends the earlier findings in important ways, including better definition of the spatial relationships between the Pt center and the BLM cleavage sites that leads to altered cleavage patterns, the demonstration that conformational alteration in DNA can lead to novel BLM-mediated DNA cleavage patterns, and the finding that platination does not alter the chemistry of DNA cleavage.

Acknowledgment. We thank Dr. Robert Hertzberg, SK&F Laboratories, for helpful discussions and for assistance with characterization of the oligonucleotides. This study was supported by PHS Grant CA38544, awarded by the National Cancer Institute, D.H.H.S., and by an American Cancer Society Scholar Grant to B.G.

Registry No. BLM A_2, 11116-31-7; BLM, 11056-06-7; *cis*-DEP, 14096-51-6; 5′-d(CGCT$_3$A$_3$CGC), 113403-23-9; Pt[d(CGCT$_3$A$_3$G$_2$)]·d-(C$_2$T$_3$A$_3$GCG), 113403-26-2.

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Additions and Corrections

Determination of the Absolute Configurations of the Isomers of Triamminecobalt(III) Adenosine Triphosphate [*J. Am. Chem. Soc.* **1986**, *108*, 4167–4171]. DAVID C. SPECKHARD, VINCENT L. PECORARO, WILSON B. KNIGHT, and W. W. CLELAND*

The assignments at the α-phosphate were inadvertently switched, so that endo isomers are really exo and vice versa. The β-phosphate assignments are correct. Thus isomers **1–4** (in order of elution from cycloheptaamylose) correspond to Λ-exo, Δ-exo, Δ-endo, and Λ-endo, respectively.

Nitroethylene Yields *N,N*-Dihydroxyiminium–Methylium Dication in Trifluoromethanesulfonic Acid. Dications Stabilized by Y Delocalization [*J. Am. Chem. Soc.* **1987**, *109*, 7036]. TOMOHIKO OHWADA, AKIKO ITAI, TOSHIHARU OHTA, and KOICHI SHUDO*

Page 7038, reference 12 (p7038) should read as follows:

In the case of planar conformations of *N,N*-dihydroxyiminium–methylium dication total energies obtained by MP2/6-31G*//4-31G and relative energies (in parentheses) are as follows: **17**, −282.6202686 au (0.00 kcal/mol); **18**, −282.6122271 au (5.05 kcal/mol); **19**, −282.603404 au (10.58 kcal/mol). (The other descriptions are deleted. This correction, however, does not make any revision in the discussion and conclusion described in the text.)