

Index

a

- ab initio* calculations 4
- based on plane wave pseudo-potential 234
- Hartree–Fock SCF method 40
- linear and nonlinear optical properties 61–79
- molecular orbital calculation method 4
- performed on LCB 266
- PWPP calculation 31, 34, 36, 72
- absorption spectroscopy 274
- accelerated crucible rotation technique (ACRT) 134
- acceptance angle 143
- ACP model. *See* anionic coordination polyhedra (ACP) model
- ACRT. *See* accelerated crucible rotation technique (ACRT)
- AFM. *See* atomic force microscopy (AFM)
- alkaline earth metal elements energy 41
- alkaline metal energy 41
- angle-tuning LBO-based OPG/OPA system 152
- angle tuning OPO systems 147
- anionic coordination polyhedra (ACP) model 227, 228
- anionic group model
 - configurations 31–33
 - energy-level electronic structure 61
 - anionic group theory 15, 16–25, 28, 37, 38, 61, 71, 75, 79, 82, 85, 87, 101, 137, 153, 270, 279, 289
 - degree of approximation 65
 - of NLO effects in crystals 16
 - for NLO susceptibility 17
 - principle 6
 - $(\text{IO}_3)^-$, lone-pair orbital 59
 - MO energy and percentage population 46
 - $(\text{B}_3\text{O}_7)^{5-}$ 50
 - $(\text{B}_5\text{O}_{10})^{5-}$ 54

- $[\text{B}_5\text{O}_6(\text{OH})_4]^-$ 54
- $(\text{H}_2\text{PO}_4)^-$ 56
- $(\text{IO}_3)^-$ 58
- $(\text{NO}_2)^-$ 59
- $(\text{SbF}_5)^{2-}$ 58
- anti π -conjugated orbital 60
- approximate quantum chemical methods 16–25
- atom-cutting analysis 75
- atomic force microscopy (AFM) 171

b

- BABF. *See* barium aluminum borate difluoride, $\text{BaAlBO}_3\text{F}_2$ (BABF)
- β - BaB_2O_4 (BBO) 117–131, 310
 - applications 127–131
 - BBO linear optical properties 120–122
 - BBO nonlinear optical properties 122–127
 - BBO single-crystal growth 117–120
 - electronic structure 44–47
- BaB_2O_4 compound
 - melting point 118
 - phases 117
- BaB_2O_4 –NaF pseudo-binary system 119
 - phase equilibrium diagram 119
- BaB_2O_4 – Na_2O system, phase diagram 118
- band gaps 35, 100, 101
- barium aluminum borate difluoride, $\text{BaAlBO}_3\text{F}_2$ (BABF) 233–245
 - atomic coordinates 235
 - conversion efficiency 245
 - crystallographic data 235
 - crystal structure 236
 - redetermination 234–237
 - effective nonlinear optical coefficient 245
 - equivalent isotropic displacement parameters 235
 - grown by MSSG technique 241
 - homogeneity plot 238

- interference diagram 240
- laser-induced damage 244
- linear and nonlinear optical properties 239–243
- optical transmission spectrum 239
- powder X-ray diffraction trace 237
- refractive indices 241, 242
- SHG output power 245
- single-crystal growth 237–239
- solubility 238
- transmission spectrum in UV–VIS–NIR region 242
- UV harmonic generation capability 244, 245
- UV transmission spectrum 241
- BaTiO₃ crystal, SHG coefficients 29
- β-barium borate (BaB₂O₄, BBO). *See* β-BaB₂O₄
- BBO lattices 48
- BBO–Na₂O phase system 119
- BBO-optical parametric oscillator device 129
 - disadvantage 130
 - photograph 131
- beam quality factor 160
 - IR source 128
- BIBO crystals 65–68, 71–74, 199, 266
 - advantages 126
 - applications 127
 - band gap 7
 - crystal structure 118
 - disadvantages 102
 - discovery 6
 - electron density contour 74
 - growth 118
 - nonlinear optical effects 73
 - photon–electron spectrum 45
 - refractive indices 66, 67, 72–74, 122
 - SHG coefficients 117, 123
 - analysis 67
 - calculated and experimental values 73
 - space structure 71
 - temperature bandwidth 126
 - transmittance on UV side 121
 - unit cell 72
 - valence band spectrum calculation 45
 - XPS spectrum 45
- birefringence phase matching 11
- bismuth triborate (BiB₃O₄, BiBO) 288
 - applications of 297
 - BiBO used for SHG 297–299
 - for sum third harmonic generation 299
 - used for OPO and OPA 300, 301
 - basic physical properties 291–293
 - B parameters and d_{eff} , comparison of 346
 - crystal growth of α-BiBO 290, 291
 - crystal structure and phases 288–290
 - layered structure 289
 - nonlinear properties 293–297
 - phase-matching configurations, and directions 295
 - temperature phase-matching bandwidths (FWHM) 296
- (BO₄)^{5–} borate group, schematic electronic energy-level structure 100
- B₂O₃–CsF flux system 216
- (B₃O₆)^{3–} group, charge density 64
- (BO₃)^{3–} group, electronic energy levels 52
- (B₃O₆)^{3–} group structure 44
 - electronic energy-level structure 48
- (B₃O₇)^{5–} group, structure 49
 - electronic energy level 51
- bond angles 225
- bond charge model 3
- bond electrons 62
- bond parameter methods 3, 22, 30
- bond valence analyses 236
- [B₅O₆(OH)₄]- anionic group structure 53
 - band gaps 100
- borate crystals 310
 - anionic group structures 11
 - comparison of B parameters and d_{eff} 346
 - nonlinear optical crystals
 - anionic group theory 16–25
 - approximate quantum chemical methods 16–25
 - β-BaB₂O₄ (BBO), electronic structure 44–47
 - band gaps 100, 101
 - BBO and LBO family crystals 65–68
 - from BBO to LBO development 102, 103–106
 - BIBO crystal 71–74
 - calculations and analysis for 65–74
 - computational methods 61–65
 - computer-assisted molecular design system for 79–87
 - discovering BBO history 101, 102
 - for frequency conversion 117–245
 - iodate crystals 29–32
 - electronic structure 57–60
 - isolated anionic group type, absorption edge calculations for 44–60
 - KBBF, BaAlBO₃F₂ (BABF) and Sr₂Be₂B₂O₇ (SBBO) family crystals 68–71
 - to KBBF crystal development 103–106

- from KBBF to SBBO family development 106–109
 - KB5 ($\text{KB}_5\text{O}_8 \cdot 4\text{H}_2\text{O} / [\text{KB}_5\text{O}_6(\text{OH})_4] \cdot 2\text{H}_2\text{O}$) crystal 36, 37
 - $\text{KBe}_2\text{BO}_3\text{F}_2$ (KBBF), electronic structure 49–52
 - $\text{KB}_5\text{O}_8 \cdot 4\text{H}_2\text{O}$, electronic structure 52–55
 - KH_2PO_4 (KDP) crystals, calculations and analysis for 77–79
 - KH_2PO_4 (KDP), electronic structure 55–57
 - LiB_3O_5 (LBO), electronic structure 47–49
 - linear and nonlinear optical properties, *ab initio* calculations 61–79
 - model and approximation 39–44
 - molecular orbital calculation method 20–25
 - molybdate crystals 33, 34
 - NaNO_2 crystals 37–39
 - calculations and analysis for 74–76
 - electronic structure 57–60
 - Na_2SbF_5 crystals 34–36
 - calculations and analysis for 76, 77
 - electronic structure 57
 - NLO borate crystals development 87–109
 - calculations and analysis for 74–79
 - devices, material requirement for 79–81
 - with molecular engineering approach 101–109
 - NLO/LO properties, basic structural units 87–93
 - perovskite and tungsten-bronze type of crystals 25–29
 - phosphate crystals 32, 33
 - second-order susceptibilities 93–99
 - theoretical basis for development 15–109
 - theoretical evaluation 82–87
 - theoretical model 16–20
 - typical NLO crystals calculation
 - with anionic group theory, SHG coefficients for 25–39
 - UV side, anionic group and inorganic crystals absorption edge 39–61
 - nonlinear optical effects in 70
 - borate series NLO crystals, discovery 15
 - boron–oxygen groups 102
 - Brewster-cut CLBO crystals 163
 - bulk KBBF crystal 182
 - conversion efficiency relationship with peak-power density 185
 - cutoff wavelength 187
 - hydrothermally grown 184
 - interference pattern 187
 - morphology 183, 184
 - phase-matching angles 190
 - phase-matching characteristics 188
 - refractive indices 189
 - right-angle prism 188, 189
 - space group 184
 - space structure 186
 - transmittance 187, 188
 - X-ray rocking curve 183
 - bulk laser-induced damage thresholds 168
 - bulk RBBF crystal 205
- c**
- Cartesian components 63
 - Cartesian coordinates 34
 - Ce:LCB, spectral properties of 265
 - centimeter-sized crystals 227
 - cesium lithium borate $\text{CsLiB}_6\text{O}_{10}$ (CLBO) 161–178, 310
 - applications 171–178
 - basic structural properties 161
 - B parameters and d_{eff} comparison of 346
 - crystalline single phase 168
 - degradation crystallinity and solution 165
 - high-quality, advanced growth technology for 165–170
 - interferogram 166
 - ion beam etching for surface damage resistance enhancement 170, 171
 - laser damage durability 172
 - lattice constants 165
 - linear and nonlinear optical properties 161–164
 - phase-matching properties 175, 177
 - photograph 162
 - projection 162
 - sample 169
 - thermooptic dispersion formulas 163
 - UV light generation, properties for 164
 - cesium triborate (CsB_3O_5 , CBO) 310
 - charge density 64, 74
 - contour 76
 - distribution 64, 76
 - charge transfer model
 - of conjugated organic molecules 3
 - CLBO. *See* cesium lithium borate $\text{CsLiB}_6\text{O}_{10}$ (CLBO)
 - CNDO method. *See* complete neglect of differential overlap (CNDO) method
 - CNDO-type approximation method 78
 - S-type approximation 38
 - coherence length, thickness 9
 - coherent anti-Stokes Raman scattering (CARS) microscopy 153
 - commercial laser systems 81

- complete neglect of differential overlap (CNDO) method 20, 22
 - approximation 23, 24
 - computational methods 61–65
 - computer-assisted molecular design system 15, 79–87, 86
 - for new NLO crystals 88
 - π -conjugated orbital system 38, 68, 94, 97
 - conjugated organic molecules
 - charge transfer model 3
 - continuous wave (CW)
 - harmonic generation 348–355
 - laser system 81
 - conventional mechanical polishing process 170
 - conversion coefficient 18
 - conversion efficiency 80, 126, 127, 130, 160, 175, 177, 183, 185, 197, 198, 200, 201, 202
 - conversion system 176, 273
 - cooling process 156
 - Coulomb correction potentials 44
 - critical phase-matched (CPM) LBO configuration 147
 - crystal cracking 315
 - in CLBO crystal 165
 - due to growth rate variation 317, 318
 - due to large supercooling 315, 317
 - crystal growth mechanism 154
 - crystallization system 224
 - crystallographic system 131, 135
 - $\text{CsBe}_2(\text{BO}_3)\text{F}_2$ (CBBF) crystal 218
 - atomic coordinates 216
 - crystallographic data 215
 - DSC curve 219
 - equivalent isotropic displacement parameters 216
 - interference pattern 215
 - measured and calculated, refractive indices 220
 - phase-matching angles, for type I SHG with 222
 - polycrystalline samples 215
 - powder XRD diffraction 217
 - structure 216, 217
 - transmittance in IR region 220
 - transmittance in UV region 219
 - type I SHG phase-matching angles *vs.* fundamental wavelength 223
 - X-ray Rietveld refinement 215
 - CsB_3O_5 (CBO) crystal 153–161
 - applications 159–161
 - conversion efficiency 159, 160
 - linear optical properties 156, 157
 - Mohs hardness 159
 - Nd-based lasers 159
 - nonlinear optical properties 157–159
 - SHG coefficients 154, 157
 - single-crystal growth 154–156
 - transparent single crystals 155
 - unit cell 154
 - UV and IR absorption edges 156
 - CW. *See* continuous wave (CW)
 - CW diode-pumped mode-locked Nd:YVO₄ laser system 152
 - CW power density 146
 - Czocharlski (CZ) method 120, 155
- d**
- damage threshold 144, 158, 193
 - dangling bonds 52, 85, 100, 101, 103
 - deep ultraviolet generation (DUV)
 - coherent light 103
 - harmonic generation 212
 - capability 194
 - laser photoemission electron microscopy (DUV-PEEM) 224
 - light sources 173, 174
 - wavelength region 161
 - density functional theory (DFT) 44, 61
 - first-principles 65
 - density matrix 21
 - DFT. *See* density functional theory (DFT)
 - dielectric function 62, 64
 - imaginary part 65
 - differential scanning calorimetry (DSC) 217
 - differential thermal analysis (DTA) 237
 - diode-pumped multilongitudinal mode Q-switched Nd:YAG laser 127
 - diode-pumped Nd:YVO₄ laser 160
 - diode-pumped solid-state laser technology 309
 - discrete variational X_α (DV- X_α) method 42, 43, 46, 49, 51, 53, 56, 58, 59
 - disk texturing 361
 - distrontium diberylo-borate $\text{Sr}_2\text{Be}_2\text{B}_2\text{O}_7$ (SBBO) 107
 - linear and nonlinear optical properties 107
 - unit cell structure 108
 - domain inversion technique 10
 - DSC. *See* differential scanning calorimetry (DSC)
 - DTA. *See* differential thermal analysis (DTA)
 - DUV. *See* deep ultraviolet generation (DUV)
 - DV-SCM- X_α approximation method 60, 85
 - DV-SCM- X_α localized cluster calculation method 15
 - DV SCM- X_α program 100

e

- EDFA. *See* erbium-doped fiber amplifier (EDFA)
 - EHMO method. *See* extended Hueckel molecular orbital (EHMO) method
 - electric field 2
 - electron–core interactions 17
 - electronics manufacturing 360
 - energy band calculation method 4
 - energy band theory 38
 - energy conversion efficiency 245
 - erbium-doped fiber amplifier (EDFA) 175, 362
 - Euler angles 84
 - exchange correction potentials 44
 - extended Hueckel molecular orbital (EHMO) method 20, 21, 24–26, 30, 32, 34, 35, 41
- f**
- FBGs. *See* fiber Bragg gratings (FBGs)
 - femtosecond Ti:sapphire laser 212
 - Fermi's golden rule 62
 - fiber Bragg gratings (FBGs) 364
 - fabrication 364
 - first-order perturbation theory 28
 - first-principles methods 15, 61
 - flashlamp-pumped Q-switched Nd:YAG laser 221
 - fluoride series crystals 77
 - flux pulling method 119, 132
 - advantages 120
 - Fock Hamiltonian 23, 24
 - four-color output light beam, experimental OPO setup for 150
 - frequency conversion
 - β -BaB₂O₄ 117–131
 - borate nonlinear optical crystals 117
 - efficiency 327
 - KBBF family 178–245
 - LBO family 131–178
 - techniques 9–11, 343, 359
 - figures of merit 343, 344, 347
 - industrial applications of 359
 - normalized conversion efficiency 343, 344, 347
 - frequency-doubled Argon ion laser 177
 - FT-IR spectroscopy 169
 - full-matrix least square methods 234
 - fullwidth at half-maximum (FWHM) 141, 181, 272, 285, 303, 307, 308
 - FWHM. *See* fullwidth at half-maximum (FWHM)

g

- gain flattening filters (GFFs) 364
- GaN semiconductor laser 10
- Gaussian 92 *ab initio* calculations 65, 77, 94
- Gaussian package 20
- GdCa₄O(BO₃)₃ 276
 - applications of crystals 282, 283
 - second harmonic generation for 283, 284
 - used for laser host crystal 284, 285
 - basic physical property of 278
 - crystal structure 275, 276
 - nonlinear properties of 279–282
 - single-crystal growth 276–278
- GdYCOB (Gd_xY_{1-x}Ca₄O(BO₃)₃) 301
 - applications 302
 - NCPM SHG for Nd:YAG Laser 307, 308
 - NCPM SHG for Ti:Sapphire laser 308, 309
 - NCPM THG for Nd:YAG laser 302–307
 - basic properties 301, 302
- generalized gradient approximations (GGAs) 61
- GFFs. *See* gain flattening filters (GFFs)
- GGAs. *See* generalized gradient approximations (GGAs)
- goniometer–spectrometer system 218
- green luminescence 156
- group velocity dispersion (GVD) 153
- GVD. *See* group velocity dispersion (GVD)

h

- Hamiltonian elements 23
- Hartree–Fock (HF) equation 21, 23, 42
- Hartree–Fock quantum chemical method 78
- heat field rotation method (HFRM) 119
- heat field symmetry control technique, development 134
- heat–mass transfer 119
- hexagonal cell dimensions 118
- HFRM. *See* heat field rotation method (HFRM)
- highest occupied ionic orbitals (HOIOs) 40, 60
- highest occupied molecular orbitals (HOMOs) 57
- high-intensity pump sources 128
- high-power diode-pumped Nd:YAG laser 160
- high-temperature flux method 181, 204
- high-temperature *in situ* observation device 227
- HOIOs. *See* highest occupied ionic orbitals (HOIOs)
- HOMOs. *See* highest occupied molecular orbitals (HOMOs)
- hydrogen bond system 55, 56

- hydrothermal method 204
- hydrothermal RBBF crystals
 - grown by seed method 206
 - grown by spontaneous nucleation 205
- i**
- ICBO crystal 161
- ICSD. *See* inorganic crystal structure database (ICSD)
- immersion-seeded solution growth (ISSG) technique 120
- infinite lattice network 202
- infrared region
 - transmittance spectrum in 121
- injection-seeded master oscillator power amplifier system 174
- inorganic crystals, absorption edges 85
- inorganic crystal structure database (ICSD) 106
- interatomic chemical bonds 76
- intra-atomic transitions 64
 - dipole transition 65
- intracavity method 10
- I-O bond parameter method 30
- iodate crystals 29–32
 - electronic structure 57–60
 - SHG coefficients 30
- ion beam-etched CLBO
 - AFM images 171
- ion–electron transport 6
- IR transmittance spectrum 169
- isolated anionic group type, absorption edge calculations for 44–60
- ISSG technique. *See* immersion-seeded solution growth (ISSG) technique
- j**
- J-O calculations 265
 - of Ce:LCB, Nd,Yb:LCB, and Pr:LCB 265
- k**
- $K_2Al_2B_2O_7$ (KABO) crystal 228
 - advantages 232
 - applications 233
 - damage threshold 232, 233
 - evolution procedure 228
 - linear optical properties 228–230
 - Maker fringes of d_{11} 231
 - morphology evolution process 227
 - nonlinear optical properties 231, 232
 - phase-matching angles 232
 - phase-matching curve 231
 - refractive indices 230
 - structure projection 224, 225
 - transmission spectrum 229
 - UV absorption mechanism 228
- KBBF-CaF₂ prism-coupling technique 189
- KBBF-prism-coupled device (KBBF-PCD) 195
 - stability test 199
- KB5 (KB₅O₈·4H₂O/[K[B₅O₆(OH)₄]₂·2H₂O] crystal 36, 37
 - absorption range 53
 - transparent spectrum 55
- KB₂BO₃F₂ (KBBF) crystal family 69, 101, 178–245
 - absorption edge 52
 - BaAlBO₃F₂ 233–245
 - CsBe₂BO₃F₂ (CBBF) crystal 213–224
 - d_{11} coefficient 194
 - electronic structure 49–52
 - frameworks 214
 - K₂Al₂B₂O₇ 224–233
 - RbBe₂(BO₃)F₂ (RBBF) crystal 202–213
 - stability test 197
 - UV transmittance spectrum on UV side 53
- KBF₄–BeO system 178
- KB₅O₈·4H₂O, electronic structure 52–55
- K₂CO₃–B₂O₃ systems 225
- Kerr effect 266
- Kerr-lens mode-locked Ti:sapphire laser 153
- KH₂PO₄ (KDP) crystal family 32, 33
 - *ab initio* calculations for 77
 - absorption behavior 57
 - atom-cutting analysis results for 78
 - calculations and analysis for 77–79
 - electronic structure 55–57
 - electrooptic coefficient 33
 - ferroelectric transition temperature 56
 - transmittance spectrum 55
- Kleinman symmetry 122, 137, 158
- knife-edge technique 171
- KTaO₃ crystal 27, 28
 - electric field-induced SHG coefficients 29
- Kyropoulos technique 155
- l**
- La₂CaB₁₀O₁₉ (LCB) 261
 - basic physical, and optical properties 263, 264
 - crystal structure 262
 - laser and optical applications 265
 - as optically operated limiters 266
 - SFD application of Nd:LCB 265
 - SHG and THG applications of LCB 265
 - nonlinear properties of 264, 265
 - La³⁺ ions 261

- Langmuir–Blodgett films 10
- laser experiments 179
- laser frequency conversion 80, 163
- laser-induced damage mechanism 164
- laser-induced damage threshold (LIDT) 164, 169, 170
- laser marking 360, 361
- LB4 crystals 309, 310
- characterization, along phase-matching directions 319
 - fourth and fifth harmonic generation of Nd:YAG 327, 328
 - laser damage 328, 329
 - linear and nonlinear optical properties 323–325
 - nonlinear optical properties 325–327
 - optical homogeneity 319, 320
 - scattering 320–323
 - sum frequency generation of 328
 - surface damage threshold 329–332
 - crystal growth 312
 - cracking problem during growth 315–318
 - thermal treatment of LB4 melt 312–315
 - crystallization electromotive force 312, 313
 - grown in phase-matching directions 318, 319
 - nonlinear properties 310, 311
 - optimum composition for growth 310, 311
 - quasi-phase-matching structure formed in 332
- LCAO self-consistent field method 23
- LDA. *See* local density approximation (LDA)
- Levine's bond charge model 34
- LiB₃O₅ (LBO) crystal family 65–68, 131–153, 178
- advantages 145
 - applications 145–153
 - cesium lithium borate CsLiB₆O₁₀ (CLBO) 161–178
 - CsB₃O₅ (CBO) 153–161
 - electronic structure 47–49
 - large-size 134
 - lattices 48, 104
 - UV transmittance in UV side 48
 - valence-band XPS spectra 51
 - LBO linear optical properties 135, 136
 - LBO nonlinear optical coefficients 136–145
 - LBO–OPO system 152
 - LBO single-crystal growth 132–135
 - MoO₃ binary system 133
 - phase-matching curves 140
 - principal refractive indices 136
 - refractive indices 66, 67
 - SHG coefficients 137, 154
 - analysis 67
 - transmittance
 - in infrared region 136
 - spectrum 135
 - on UV side 135
 - unit cell structure 132
 - X-ray powder diffraction data 132
- LIDT. *See* laser-induced damage threshold (LIDT)
- linear combination of atomic orbitals (LCAO) 21
- linear expansion coefficients 21
- linear optical (LO) 1
- refractive indices 61
- linear polarizability 16
- linear refractive indices 70
- linear susceptibilities, anisotropy of 85
- Li₂O–B₂O₃ system
- nucleation thermodynamics 133
 - phase diagram 131, 132
 - viscosity in 133
- Li₂O–MoO₃ flux system 134
- lithium tetraborate (Li₂B₄O₇). *See* LB4 crystals
- lithium triborate (LiB₃O₅, LBO) 310
- B parameters and d_{eff} , comparison of 346
- LO. *See* linear optical (LO)
- local core matrix 24
- local density approximation (LDA) 61, 63
- localized spontaneous nucleation 180
- lone-pair bonds 34
- lone-pair electron 73
- long period gratings (LPGs) 364
- Lorentz linewidth modification 49
- lowest unoccupied ionic orbitals (LUIOs) 40
- LUIOs. *See* lowest unoccupied ionic orbitals (LUIOs)
- m**
- Madelung correction 20, 24
- Madelung energy 47
- Madelung potential energy corrections. *See* Madelung correction
- Madelung's potential 40, 41, 43
- Maker fringes
- of d_{11} coefficient 223
 - technique 36, 123, 137, 211, 221, 231, 243
- master oscillator power amplifier (MOPA) 160, 174, 175
- mercury laser system 273
- metrology 361–363
- microscopic second-order susceptibilities 87, 95, 96, 98
- (B₅O₁₀)⁵⁻, [B₅O₆(OH)₄]⁻, (B₄O₃)⁶⁻ 96
 - coefficients 106

- planer six-member ring molecules 96
 - microscopy 361–363
 - Millennium Research for Advanced Information Technology (MIRAI) Project 176
 - Miller's rule 2, 128
 - minimum deviation method 229, 241
 - mixing-in-aqueous-solution techniques 168
 - modified middle-seeded solution growth (MSSG) method 225
 - Mohs hardness scale 211, 222
 - molecular configurations 89–91, 98, 99
 - molecular engineering approach 39, 79
 - molecular orbital calculations
 - method 20–25
 - CNDO-type approximation 21–24
 - EHMO-type approximation 24–25
 - for nonplanar molecules 23
 - molecular orbital transition moments 19
 - molybdate crystals 33, 34
 - molybdenum oxide flux 134
 - momentum matrix elements 63
 - (MO₆)ⁿ⁻ coordination octahedron, deformation modes 26
 - monolithic wavelength converter 305–307.
 - See also* GdYCOB
 - output power of third-harmonic wave, measurement 306
 - using GdYCOB and KTP 305
 - MoO₃-based flux, advantages 133
 - MOPA. *See* master oscillator power amplifier (MOPA)
 - MSSG method. *See* modified middle-seeded solution growth (MSSG) method
 - muffin-tin approximation 42
 - multiphoton absorption process 145
 - multiple scattering-X_α (MS-X_α) method 42
- n**
- NaF flux system 237, 238
 - NaF pyrohydrolysis 119
 - NaNO₂ crystals 37–39
 - calculations and analysis for 74–76
 - electronic structure 57–60
 - linear and nonlinear optical coefficients 74
 - SHG calculation 59
 - SHG coefficients in 85
 - nanosecond pulsed Nd:YAG laser 195
 - Na₂SbF₅ crystal 34–36
 - atom-cutting analysis for 76
 - calculations and analysis for 76, 77
 - charge density contour 77
 - electronic structure 57
 - SHG coefficients 34
 - NCPM. *See* noncritical phase-matching (NCPM)
 - Nd-based laser systems 81, 171, 179, 213, 233
 - harmonic generation 127, 195, 220
 - Nd³⁺-doped LCB (Nd:LCB) 261
 - Nd:GdCOB, as practical SFD crystal 286–288
 - effective NLO coefficients 286
 - infrared laser properties 286, 287
 - maximum green output power 286
 - optical conversion efficiency 287
 - optimal doping 287
 - output and pump power, relationship between 287
 - output stability 288
 - pump-absorbed power 286
 - Sellmeier equations 286
 - spectroscopic properties 286, 287
 - Nd:LCB
 - as-grown 263
 - measurements 265
 - SFD application of 265
 - Nd:YAG laser 129, 138, 140, 158, 163, 228, 233, 301, 310
 - fourth harmonic generation 183
 - frequency
 - second-harmonic and sum frequency generation 158
 - generation 80
 - harmonics 141
 - phase-matching coupling 124
 - Q-switched and rotating mirror 123
 - second harmonic and sum frequency generation 124
 - tuning curves 125
 - Nd:YLF laser 147, 152
 - Q-switched Nd:YLF laser 158
 - near-hexagon trigonal symmetry 181
 - neodymium lasers, harmonic generation
 - phase matching configurations of 345
 - third harmonic of 359, 360
 - (NO₂) group, lone-pair orbital 38
 - noncritical phase-matching (NCPM) 81, 127, 141, 144, 146, 147, 152, 153, 302, 303, 307, 308, 309
 - nonlinear coefficients 9, 159, 294, 354
 - nonlinear optical (NLO) crystals 213. *See also* GdCOB; YCOB
 - active group theory 25
 - birefringence in 86
 - borate crystals
 - development with molecular engineering approach 101–109
 - history of development 4–7
 - calculations and analysis for 74–79

- coefficients 163
 - calculation with anionic group theory, SHG 25–39
 - IEEE/ANSI standard 122, 136
 - developments in borate series 87–109
 - devices
 - material requirement for 79–81
 - experimental and calculated absorption edges 60
 - frequency conversion
 - crystals, conditions for 8
 - higher efficiency methods 8
 - history of NLO crystals 7–11
 - macroscopic properties 2
 - NLO and LO properties 15
 - optical parameters 4
 - physical origin 3
 - property 165
 - second harmonic generation, frequency conversion efficiency 7, 8
 - SHG coefficient 4
 - structure–property relation (*See* Miller’s rule)
 - and techniques, frequency conversion history 9–11
 - theoretical understanding history 1–4
- o**
- odd-ordered crystal field 27
 - off-diagonal core matrix elements 22
 - OPO device. *See* optical parametric oscillation (OPO) device
 - optical axis system 137
 - optical coefficients 64, 74, 76, 79, 144, 281, 348, 353
 - optical contact prism-coupling device 223
 - optical devices, characterization of 355
 - finesse measurement 358, 359
 - photothermal interferometry 355
 - resonator measurement 355–358
 - optical disk standards 363
 - optical parametric oscillation (OPO) device 128, 152
 - optical-to-optical conversion efficiency 146
 - orthorhombic system 157
 - oscillating temperature regime 120
- p**
- Pariser’s calculation 18
 - PCBs. *See* printed circuit boards (PCBs)
 - Perkins-Elmer spectrophotometer 241
 - perovskite-bronze type of crystals 25–29
 - niobate crystals 25–27
 - SrTiO₃, BaTiO₃, KTaO₃ crystals 27–29
 - phase equilibrium diagram 103
 - phase-matched (PM)
 - angles 138, 141, 188, 209
 - vs. fundamental wavelength 191
 - characterization 137, 139, 142, 143
 - method 9, 36, 81, 123
 - range 5, 49, 81, 90, 101, 102, 138, 146, 192, 209
 - SHG properties 265
 - phosphate crystals 32, 33
 - photomultiplier tube (PMT) 221
 - photon energy-tunable photoemission spectrometer 202
 - plane wave pseudopotential (PWPP) total energy package 15
 - PM. *See* phase-matched (PM)
 - PMT. *See* photomultiplier tube (PMT)
 - point charge model 27
 - point-group symmetry 22
 - potassium difluo-diberylylo-borate KBe₂BO₃F₂ (KBBF) 104
 - framework 106
 - linear and nonlinear optical properties 107
 - unit cell 105
 - powder SHG test 117
 - PPLN devices 10
 - printed circuit boards (PCBs) 360
 - prism-coupling technique 195, 212
 - Pr:LCB 262, 265
 - proton transfer potential energy curve 77
 - pseudopotential method 38, 72
 - pseudo-ternary system 180
 - KBBF primary crystallization 180
 - pulsed OPO system 130
 - PWPP method 29, 68, 75, 78, 79, 82
- q**
- Q-switched diode-pumped Nd:YAG master oscillator 173
 - Q-switched Nd:YAG laser 244
 - Q-switched Nd:YLF laser 158
 - Q-switched Nd:YLF oscillator 145
 - quantum chemical approximation method 39
 - quantum chemistry calculation methods 17
 - quartz crystal, second harmonic generation (SHG) 1
 - quasi-cw DUV sources 176
 - quaternary phase diagram 180
- r**
- Raman spectroscopy 44
 - rare earth (RE) elements 261
 - RbBe₂(BO₃)F₂ (RBBF)
 - building units 202

- crystal structure 203, 204
 - d_{11} coefficient 211, 212
 - disadvantages 211
 - frameworks 214
 - interference pattern 203
 - measured and calculated refractive indices 207
 - phase-matching angles for type I SHG with 208, 209
 - RBBF-PCD device 212, 213
 - special prism-coupling device, schematic presentation 212
 - transmittance
 - in IR region 207
 - in UV region 206
 - type I SHG phase-matching angles vs. fundamental wavelength for 210
 - Rb₂Be₂Si₂O₇ unit cell structure 107
 - real-space atom-cutting 72
 - analysis 69, 74
 - techniques 63, 70, 78
 - ReCOB family
 - development of 266, 267
 - ternary phase diagram 277
 - refractive indices 66, 207, 218, 264
 - calculated and experimental values 69
 - data 120
 - dispersion 192, 210
 - dispersion curve 221, 230
 - experimental values 157
 - RE:LCB crystals
 - basic physical and optical properties 263, 264
 - nonlinear properties 264, 265
 - synthesis and crystal growth 261, 262
 - resonance integral 22
 - Rietveld analysis 214
 - Rigaku Thermoflex TAS 200 237
 - Roothaan equations 21
 - Roothaan–Hartree–Fock (RHF) equation 40
- s**
- saturation effect 28
 - SAW devices. *See* surface acoustic wave (SAW) devices
 - scattering process 17
 - Schrodinger equation 43
 - second harmonic generation (SHG) 1, 80
 - coefficients 4, 8, 15, 17, 26, 28, 57, 62, 68, 70, 83, 84, 93, 117, 122
 - atom-cutting analysis for 78
 - calculation 31, 37
 - conversion efficiency 146
 - crystals, applications 62
 - powder SHG test technique 3, 4
 - type I phase-matching angles 207
 - second-order perturbation theory 18, 39
 - second-order susceptibilities 20, 37, 63, 82, 88, 93–99, 94
 - calculation 93
 - tensor 97
 - seeded growth technique 180
 - seed method 205
 - seed-submerged growth technique 156
 - self-fluxes systems 225
 - self-frequency doubling (SFD)
 - application of
 - basic physical 263
 - crystal structure of 262
 - nonlinear properties 264
 - optical properties 263
 - optical quality 262
 - SHG and THG applications 266
 - crystal 261, 278, 281, 283, 286
 - in Nd:YCOB 274
 - in ReCOB family Nd:GdCOB 267
 - Sellmeier equation 120, 124, 135, 138, 157, 161, 187, 189, 191, 192, 193, 207, 209, 211, 219, 220, 230, 231, 243, 294, 324, 325
 - SFD. *See* self-frequency doubling (SFD)
 - SFG method. *See* sum frequency generation (SFG) method
 - SHELX-97 system 234
 - SHG. *See* second harmonic generation (SHG)
 - Siamese-twin double six-member
 - molecular configurations 92, 93
 - simplified phase diagram 181
 - single-pass conversion 347
 - single-resonance sum frequency mixing cavity 177
 - single-site orbitals (SSOs) 43
 - potential 47
 - solid reaction method 132
 - solution-stirring technique 167
 - space structure resolution process 202, 213
 - spectral bandwidth 144
 - spontaneous nucleation technique 204
 - SS-TSSG method 167, 171, 173
 - stability test 195, 197–199
 - stacking fault model 184
 - stereolithography 359
 - strontium boron beryllium oxide (Sr₂B₂Be₂O₇, SBBO) 310
 - structure–property relationship 5

- sum frequency generation (SFG) method 7,
104, 124, 138, 158, 175, 233, 265, 296,
299, 328
- sum frequency mixing (SFM) 138, 152,
177, 345
- surface acoustic wave (SAW) devices 310
- t**
- temperature bandwidth
– experimental and calculated values 144
- temperature-tuned NCPM 142
- temperature tuning curve 151
- tetra-LBO 309
- TGA method 225
- thermal-optical coefficients 126, 144
- time-dependent perturbations, Fermi's
golden rule 62
- Ti:sapphire-based laser systems 128, 147,
150, 198, 301
– harmonic generation 196, 198, 200
– power output curves 201
– vs. fundamental wavelength 213
– wavelength range 199
- top-seeded solution growth (TSSG)
method 118, 119, 120, 133, 134, 155, 161
– solution-stirring technique 167
- top-seeding high temperature flux
method 179
- total power density 244
- transformation formulas 84
- transition matrix element calculation 25
- transmittance spectrum 120, 206
- traveling solvent zone melting (TSZM)
method 120
- trimming 361
- TSSG method. *See* top-seeded solution growth
(TSSG) method
- TSZM method. *See* traveling solvent zone
melting (TSZM) method
- tungsten-bronze type crystals 5, 25–29
– niobate crystals 25–27
– SrTiO₃, BaTiO₃, KTaO₃ crystals 27–29
- type II NCPM LBO–OPG, temperature tuning
curve 151
- type I NCPM LBO crystal, calculated
temperature-tuning range 148, 149
- type I SHG phase-matching angles 207
- u**
- UV-fused silica prism 195
- UV-NLO crystals 39, 44
– development 6
- UV solid-state laser system 173, 310
- v**
- vapor deposition method 132
- vapor transport equilibration (VTE)
process 156
- VASP program. *See* Vienna *ab initio* simulation
package (VASP) program
- via hole drilling 360
- Vienna *ab initio* simulation package (VASP)
program 108
- virtual excited schemes, kinds of 20
- virtual hole processes 63
- VTE process. *See* vapor transport equilibration
(VTE) process
- w**
- wavelength division multiplexing
(WDM) 364
- wide-aperture laser systems 135
- Wigner–Eckart theorem 26
- Wigner's irreducible tensor representation
method 83
- x**
- XeCl laser 129
- X-ray diffraction (XRD) 165, 225, 234
- X-ray structural analysis 30
- y**
- Yb:LCB 265
- YCOB (Ca₄YO(BO₃)₃) 266
– crystal structure 267
– major applications 272
– self-frequency doubling 274, 275
– SHG of Nd:YAG laser radiation
272–274
– THG of Nd:YAG laser radiation 272
- optical properties, linear and
nonlinear 267–271
– dispersion relations 269
– d tensor 269
– Kleinman symmetry 269
– nonlinear optical coefficients 270
– parameters of dispersion
equations 269
– principal linear thermal expansion
coefficients 271
– thermal conductivities 271
– thermooptic dispersion 271
- ReCOB family, development of 266, 267
– structural properties 267, 268
- z**
- zero differential overlap approximation 22

